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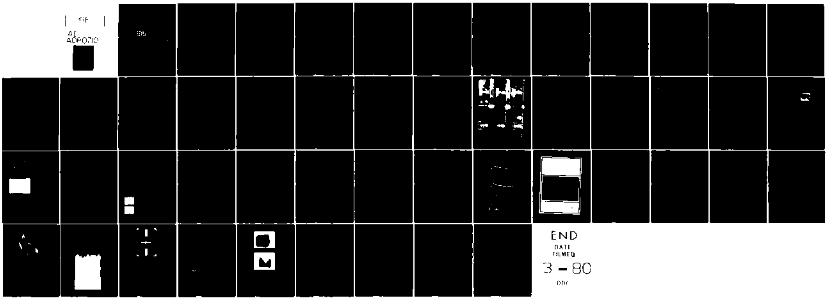

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A submillimeter heterodyne receiver/radiometer has been developed that covers the spectral range from 100 μm to 1 mm with NEPs of 10^{-19} W/Hz or noise temperatures of 7000°K. It uses GaAs Schottky diodes as mixers in quasi-optical configurations. Tunable far IR radiation has been generated and used to carry out double resonance spectroscopy experiments in CH_3F. Planar mixer diodes have been used to analyze the spectral properties of far IR lasers. Various applications of this newly developed near-mm technology have been explored.

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FAR INFRARED AND SUBMILLIMETER TECHNOLOGY

FINAL REPORT

P. E. TANNENWALD

10 JANUARY 1979

U. S. ARMY RESEARCH OFFICE

PROPOSAL 13987-P

LINCOLN LABORATORY
MASSACHUSETTS INSTITUTE OF TECHNOLOGY

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I. Introduction

During the past several years the heightened interest and consequent increased activity in the submillimeter field have brought substantial advances. Potential applications, military as well as scientific are now closer to realization. These applications include radio astronomical measurements, plasma diagnostics, frequency standards and spectroscopy, atomospheric propagation, aeronomy, high-altitude aircraft- and satellite-based surveillance, and point-to-point detection and image/scanning systems.

Lincoln Laboratory's contribution has significantly changed the state of technology in submillimeter detectors. We have improved the sensitivity of Schottky diode heterodyne detectors from an NEP of 10^{-16} W/Hz to 10^{-19} W/Hz, and the receiver/radiometer DSB mixer temperature to 4200°K throughout the submillimeter region. This gain in over three orders of magnitude in sensitivity has had a significant impact on the entire field, particularly since we have made an effort to widely distribute the technology and to collaborate or coorporate with other groups in the undertaking of new experiments.

In the process of exploring the applications of this technology, the problem now outstanding in the submillimeter region has become apparent, namely the availability of sources. We propose to apply Lincoln Laboratory's unique capabilities to the development of tunable submillimeter sources suitable for both field applications and scientific research. These sources are to be moderatively powerful (in the 1 to 10 mW range) and suitable for use as local oscillators as well as new classes of spectroscopic experiments that depend on more power than has previously been available.

The long-term program plan is to phase out optically pumped lasers. Although they are currently the workhorse in the field, they are too unstable, too bulky and too restrictive in the frequency domain. The sources we want to develop will be either completely solid state or hybrid systems using powerful mm sources such as extended interaction oscillators (EIOs) or carcinotrons; these tubes will be used in conjunction with solid state components for harmonic generation.

Two of our approaches are based on technologies which we understand and which will have a high probability of success. The first of these involves a semiconductor device called a TUNNETT, which will be described in detail below. The second approach makes use of our planar diodes. Individual diodes can handle high mm powers without burnout. Thus, when fabricated into arrays, it should be possible to illuminate a large number of diodes with our 10 W, 140 GHz EIO and obtain substantial power in the submillimeter.

Although the detector problem has been practically solved, there remain many exciting developments in this area with the application of planar technology. For example, we have demonstrated that surface-oriented, planar Schottky diodes of the same type used in submillimeter mixing are particularly suitable for observing the mixing of two optical waves under reverse bias conditions. The immediate implication is that a much simplified and reliable frequency chain can be established between light waves and microwaves.

Planar technology has yet to be exploited for applications to monolithically integrated antenna/mixer/FET structures, perhaps cooled to low temperatures, to yield the lowest achievable noise temperatures in compact, rugged configurations.

Concurrent with the development of these new solid state sources and detectors we propose a number of demonstration scientific experiments. The purpose of these experiments will be both to delineate the capabilities of the newly developed devices and to open up new areas of scientific investigation. These experiments involve spectroscopy of atomic and molecular beams, saturation spectroscopy, and optical and submillimeter wave mixing.

II. Results to Date

Progress during the contract period has been reported in the semiannual progress reports to ARO. Below are listed the significant highlights; the details appear in the reprints which are appended to this report.

1. Radiometer/Receiver - A quasi-optical submillimeter radiometer has been developed which has an NEP of 1×10^{-19}W/Hz and a mixer temperature of 4170°K (DSB). This radiometer/receiver is becoming the basic detection system in a wide range of submillimeter investigations, including plasma diagnostics, radio astronomy, and laboratory and space spectroscopy. The corner-reflector mixer which has resulted from this research program has been widely distributed to other laboratories, and its figure of merit is now used as the design standard for mixer capability in the submillimeter region.
2. Planar Diodes - Planar, surface-oriented Schottky diodes, in which both terminals of the rectifying junction lie on the same surface, have been fabricated and used as heterodyne detectors and harmonic mixers at frequencies up to 900 GHz. These planar diodes have been put to use in applications in which their greater physical and electrical ruggedness, compared to whisker-contacted diodes, is a significant advantage.

3. Laser Physics - The planar diodes were used in an experiment in which the frequency output of a high-power, pulsed far-infrared laser was analyzed by heterodyning the laser signal with a klystron harmonic. The resultant IF was fed into a surface-acoustic-wave dispersive delay line which converted the pulse into a frequency vs time display. By virtue of this real-time spectral analysis on each laser pulse, the contributions of three narrow longitudinal modes to a 385 µm D_2O laser spectral linewidth were resolved. Furthermore, confirmation was obtained that the lasing mechanism is via a stimulated Raman process which tunes linearly with pump frequency (provided by a pulse CO_2 TEA laser).

4. Generation of Tunable Radiation - The quasi-optical Schottky diodes have been developed into tunable sources of submillimeter radiation. In one scheme, harmonics are readily generated from a tunable microwave signal introduced into the bias connection to the diode; in a second scheme sidebands are radiated from the mixer when the diode is illuminated by a laser and a tunable microwave signal is simultaneously introduced into the bias line. These techniques provide an alternative to classical harmonic generation experiments, as well as an extension to shorter wavelengths (100 µm). Given the broad IF capabilities of the mixer (greater than 2 cm^{-1} using both sidebands) and the variety of far infrared laser lines available as LOs, virtually any wavelength region can be studied.

5. Submillimeter Spectroscopy - With the use of the tunable submillimeter sources, a series of spectroscopic experiments has been undertaken in a

number of molecular gases. Besides simple absorption measurements, infrared-submillimeter double-resonance experiments have been carried out using a CO_2 laser to excite vibrational levels in the molecule. Furthermore, with pulsed CO_2 excitation, the submillimeter receiver is fast enough to allow observation of rapid radiative and collisional relaxation processes among rotational levels. In this way the excited-state rotational spectra and kinetics of CH_3F were studied. This double-resonance technique is applicable to a wide class of molecules, and is particualarly important for those systems which are candidates for optically pumped submillimeter lasers.

6. High-Altitude Submillimeter Radiometry - A submillimeter radiometer has been used in conjunction with a space simulation chamber at Lincoln Laboratory to make absorption measurements of an expanding jet of water vapor against a blackbody background. These exploratory experiments, made with BMDATC support, are designed to simulate signatures from rocket plumes as might be detected from space-based surveillance platforms.(1) They also serve to gain experience for future blackbody aeronomy experiments (such as limb sounding against the sun) to study the concentration of atmospheric constituents and their spatial and temporal distributions.

III. Proposed Program

Specific program items that can be envisioned now are discussed below; other directions may be pursued as new ideas and oportunities arise.

1. Tunable Submillimeter Sources
   a. GaAs TUNNETTs
   b. GaAs planar diodes - harmonic generation from arrays
   c. exploration of InP sources

2. Advanced Detectors

   a. complete heterodyne package with solid state LO, $T_S \sim 2000°K$ at 1 mm and NEP $\sim 2 \times 10^{-20}$ W/Hz throughout the submillimeter region

   b. cooled diode mixers

3. Monolithic Mixers Using Planar Diodes, Antennas and FETs Integrated on One Chip

4. Atomic and Molecular Spectroscopy and Far Infrared Quantun Electronics Experiments

TUNNETT Diodes as Millimeter-Wavelength Devices

Reverse breakdown in a diode may occur because of an avalanche process, a tunnel process, or a mixed avalanche-tunnel process. Microwave power may be generated by a transit-time device in reverse breakdown from either process. The common IMPATT is inherently noisy but has a relatively large RF power output in the centimeter-wavelength region for GaAs devices and in both the millimeter-wavelength and centimeter-wavelength region for Si devices. The TUNNETT (tunnel transit-time)[2,3] device will be useful as a low-noise amplifier, medium-power oscillator, self-oscillating mixer, and detector, particularly at millimeter wavelengths. There are also indications that the use of tunneling current will allow oscillation of GaAs devices above 100 GHz, which will be useful for applications in millimeter-wavelength monolithic integrated circuits. It has been shown theoretically that a very quiet TUNNETT mode device can be constructed at 100 GHz in GaAs with a predicted efficiency of 6 percent. The MITATT (mixed tunnel-avalanche transit-time) device should exhibit a noise performance vs output power tradeoff which will give a useful flexibility for designing an optimum source in an integrated system.

Figure 1 shows the device efficiency as a function of frequency for uniformly doped transit-time devices. In general the frequency of operation increases as the doping increases. Figure 1 is based on the state-of-the-art results as reported in the literature and a few estimates. It should be noted that for Si devices the efficiency is relatively constant as a function of frequency up to around 100 GHz. For GaAs IMPATT devices the efficiency is larger than for Si devices at X-band, but drops off quickly such that for frequencies above 80 GHz it is no longer suitable as a source. This rapid decrease of high frequency GaAs IMPATT performance is believed to be caused by the inability of the impact ionization process to respond to the rapidly changing electric field in GaAs. If this is the case, then GaAs transit-time devices will again operate when the electric field is high enough to cause a significant tunnel current since tunneling is an extremely fast process. Significant tunnel current can only exist when the doping is quite large ($N_d \sim 5\text{-}10 \times 10^{17} cm^{-3}$), which leads to operation at frequencies above 150 GHz for uniformly doped material. This leaves a frequency band in which uniformly doped GaAs transit-time devices will fail to operate. These effects are shown in Figure 1, where TUNNETT efficiencies have been estimated. It is possible to build GaAs transit-time devices which operate in the 80-180 GHz region by using precise doping profiling. It should be noted that not only will tunnel current allow GaAs transit-time device operation above 100 GHz, but the tunnelling mechanism will result in a very low noise device with many potential applications.

GaAs TUNNETT devices have already been constructed at Lincoln Laboratory. Uniformly doped Schottky-barrier devices can now be routinely fabricated with predicted frequency operation above 200 GHz. The dc

characteristics indicate that the mesa devices are in full tunnel breakdown. Ion implanted (high-low) GaAs Schottky-barrier devices have also been constructed for operation below 200 GHz. For these two classes of devices, considerable doping-profile optimization will be necessary in order to achieve the desired performance at each frequency range of interest.

The packaging of millimeter-wavelength devices also is a very difficult problem. The device package, usually a quartz standoff configuration, has severe mechanical and electrical requirements. A number of different package configurations have been successfully demonstrated. Since the package forms the majority of the local circuit environment of the device, considerable optimization of the package parasitic reactances will be necessary to achieve maximum performance.

Preliminary microwave oscillator circuit designs are now complete and are presently being machined. The circuits under consideration at this time are full-height waveguide with a RF bypass choke arrangement and with insertible posts or top hats. The posts and top hats will form the remainder of the local circuit environment for the device. Again, considerable optimization of the oscillator circuit will be necessary.

The first thrust of the RF testing for these TUNNETT devices will be as low-noise oscillators in the 100 to 200 GHz region. These devices will also be used in the mixer and self-oscillating mixer configuration.

InP Devices

Semiconductor device technology has been perfected to a high degree in two materials - Si and GaAs. For our particular interest, namely detectors and sources in the near millimeter region, GaAs has received the most attention. However, it now appears that InP, which is another III-V semiconduc-

tor material, has electronic properties very similar to those of GaAs. An analysis of the device physics suggests that for InP IMPATTs one can expect higher output power, higher efficiency, higher frequency operation and lower noise. The same arguments will apply to the characteristics of TUNNETT devices. Possibly the greatest advantage will hold for high peak power oscillators.

Although the program proposed here does not involve development of semiconductor technology, it could well capitalize on the activities going on at Lincoln Laboratory in InP growth, characterization and device fabrication. Thus we are proposing to explore the possibilities of InP oscillators and mixers for the millimeter and submillimeter regions; the promise of major developments in this area seem sufficiently great that we can expect to make significant contributions to systems applications as well as in physics experiments.

Cooled Mixers

A considerable reduction in system noise temperature can be obtained by cooling a millimeter or submillimeter wavelength mixer.[4,5] However, the mixer must be designed to withstand the relative thermal contraction effects in cooling from room temperature and must be able to accomodate repeated temperature cycling from room temperature to the cryogenic operating temperature with no degradation in performance. In this regard monolithic surface-oriented mixers would be expected to be superior to the quasi-optical corner-reflector mixers in which the Schottky diode is contacted by a 4λ-long tungsten whisker antenna. Corner-reflector mounts have survived cooling cycles from 300°K to 16°K, and diode I-V characteristics have been measured at 16°K, 77°K and 300°K. The behavior of the I-V characteristic as a function of temperature is dependent upon the impurity carrier concentration in the epitaxial layer, which determines whether the

conduction mechanism is field emission, thermionic field emission or thermionic emission. A plot of V_0 vs kT/q (V_0 is the voltage across the diode, q is the electronic charge) allows one to determine the dominant carrier transport mechanism in the Schottky barrier diode.

Viola and Mattauch[6] have calculated the decrease in receiver noise temperature to be expected when cooling GaAs Schottky barrier mixer diodes and the influence of carrier concentration on NEP. Carrier concentration should be in the low 10^{17} cm^{-3} range, and little improvement can be expected by cooling below 77°K. Theoretical equivalent noise temperatures for a diode having 3×10^{17} cm^{-3} carrier concentration at 330°K, 77°K and 16°K are 175°K, 70°K and 60°K, respectively. Clearly, only a slight improvement will be obtained by cooling to 16°K, rather than to 70°K, and no further improvement can be obtained by cooling below 16°K.

A vacuum chamber has been designed and built for a closed cycle helium refrigerator which allows both a short focal length focusing mirror and the diode mixer to be mounted within the vacuum chamber. The diode mixer is attached to the cold finger and can be cooled to 16°K. The diode mixer and focusing mirror combination is aligned and optimized at room temperature and the system noise temperature is measured. The vacuum chamber is closed and evacuated, the mixer is cooled to 16°K and the system noise temperture is remeasured. Corner-reflector mixer mounts will be evaluated with diode chips having a range of carrier concentration and epitaxial layer thickness. Surface-oriented integrated mixer modules will also be evaluated as cooled mixers. The initial cooled mixer measurements are being made at a wavelength of 0.4 mm. Measurements will also be made on cooled mixers operating at 3 mm and 1 mm wavelengths.

A further reduction in system noise temperature can be obtained by cooling the IF amplifier. Experiments will be made to cool mixer-IF

amplifier combinations in a closed-cycle helium refrigerator with the object of developing low-noise cooled receivers over the 3 mm to 0.1 mm wavelength range.

Finally, cooled Schottky diodes have been shown to have a substantially higher nonlinear characteristic than room-temperature diodes. This will be an advantage if cooled diodes can be used in some of the planned harmonic generation experiments.

Integrated Mixer Module for Submillimeter Wavelength Operation

A GaAs monolithic integrated circuit capable of receiving radiatively coupled submillimeter wavelength radiation has been fabricated. The radiation is received by a slot antenna and coupled to a surface-oriented mixer diode by an appropriate section of coplanar transmission line (see Fig. 2a). An integrated bypass capacitor completes the mixer circuit by providing a low impedance to RF frequencies and a high impedance to IF frequencies. The design of the planar slot antenna is complicated by the presence of the high-resistivity GaAs. Free-space radiation patterns, even from simple metal antennas fabricated on the high-resistivity GaAs, show complex lobe structure unacceptable for most applications. Therefore, the module of Figure 2(a) has been designed so that the radiation is coupled through the high-resistivity GaAs substrate. The radiation pattern obtained at 350 GHz is shown in Figure 2(b). It is nearly identical to that obtained from a frequency-scaled model at 5 GHz. These patterns were obtained in both cases by measuring the video response of the surface-oriented diode as the direction of the incident radiation was changed. The theoretically expected radiation pattern from a half-wavelength dipole is also shown in Figure 2(b) for comparison. Though the

measured radiation pattern does contain some interference from the coplanar transmission line feed, it allows the near millimeter wavelength radiation to be radiatively coupled to the device. A chip on which a number of these integrated devices have been fabricated is shown in Fig. 3.

A 2 μm diameter, circular Schottky barrier diode was used in the early integrated mixer modules. Although circular Schottky barriers give excellent performance in conventional whisker-contacted chip diodes, the circular geometry has some drawbacks when used for surface-oriented diodes. First, current collection by the surface ohmic contact adjacent to the Schottky barrier is only efficient over about half the diode periphery, and thus the diode series resistance is greater than for a conventional diode. Second, alignment of a proton bombardment mask is very critical, and a small misalignment can cause large changes in effective diode area.

Line or stripe-geometry Schottky barriers overcome the drawbacks associated with the circular goemetry. The periphery-to-area ratio of rectangular stripe diodes is greater than that of circular diodes of equal area, resulting in lower diode series resistance. The current collection efficiency of the ohmic contact surrounding a rectangular-stripe diode is much greater than for a circular diode, which also contributes to a lower series resistance. Alignment of a proton bombardment mask is less critical, and a slight misalignment causes little change in the effective diode area.

A new mask set for an integrated mixer module at 350 GHz having rectangular-stripe Schottky barrier diodes has been made. The masks produce diodes with dimensions of 2.9 x 0.6 μm and 2.5 x 1.3 μm, and use conventional 10:1 optical projection lithography. Devices are now in the process of fabrication. Electron beam lithography will be used in conjunc-

tion with the existing mask set to generate line or stripe Schottky barrier diodes with dimensions in the range 10 x 0.2 μm to 5 x 0.2 μm, which should give greatly improved performance in the submillimeter region.

Integrated mixer modules will be frequency scaled and fabricated at 3 mm (100 GHz) and at 0.4 mm with the object of developing integrated mixer modules over the 3 mm to 0.1 mm wavelength range. An FET IF amplifier will also be integrated with the mixer module once the individual components are fully characterized. An integrated 3 x 3 imaging array will be fabricated and tested when the fabrication sequence is fully evaluated and high yield is obtained.

Monolithic Arrays

Once the integrated mixer module (slot antennas and planar diodes integrated on one GaAs chip) has been perfected, arrays of these integrated antenna/diode modules with appropriate configurations, e.g., end-fire or broadside, will be fabricated. Among other factors, the spacing between these devices will determine the radiation patterns. Such an array will be able to withstand much higher incident signal powers than a single whisker-contacted diode - firstly because of the absence of a point contact, and secondly because the power is distributed among a large number of diodes. Thus we can expect to illuminate the array with an available 10 W EIO tube at 140 GHz and generate substantial tunable output at the second and higher harmonics.

Atomic and Molecular Spectroscopy and Far IR Quantum Electronics Experiments

1. In other spectral regions atomic and molecular beams have been used for spectroscopic sub-Doppler studies on specially prepared states. Until now it has not been feasible to extend these measurements into the submillimeter region because of power, linewidth and tunability constraints.

As part of a related frequency-standards effort we have recently constructed a suitable beam source. Therefore, using these new sources, we propose to examine fine-structure transitions in Mg, O^+ and other atomic systems. We will also examine multiple beam effusive sources of polar molecules in the first of a class of very high resolution submillimeter studies. Some of these experiments detect the submillimeter radiation in the visible and hence have analogies with recent studies of infrared up-conversion in metal vapors. (It is likely that some of these fundamental investigations will have practical implications.)

2. With the power levels of 10 mW anticipated in this effort it becomes reasonable to look into saturation spectroscopy in the 100 μm to 1 mm region. Both solid state systems (for example shallow donors in semiconductors) or gases (Rydberg state) are potential candidates. At first we propose using the tunables sources merely to probe levels which are saturated via other fixed-frequency sources. However, as our work proceeds, we propose using the tunable source themselves for the saturation of particular systems. In additon to obtaining information on natural linewidths of systems we can also look into stabilization and locking of sources to Lamb dips in the rotational transitions of gases.

3. In preliminary experiments we have noted that our planar diodes responded well to the visible radiation. When one considers that the light is injecting carriers into the depletion region of the GaAs diode, this effect is not at all suprising and has been seen in both FETs and IMPATTs. One can also look at the beat note between two visible lasers. If the frequency difference is below about 3000 GHz, currents will flow (just from square-law detection) which can mix with currents generated by illu-

minating the diode with submillimeter radiation. Our system has already been tested at 8 GHz using modes from an argon ion laser. Similar work at the Max-Plank-Institute for Radio Astronomy at Bonn has already gone to 80 GHz. We project our system to be able to mix visible lasers separated in frequency up to 100 cm^{-1}. These experiments have, of course, implications for our measurements of atomic and molecular systems. More importantly, they may permit a correlation calibration of visible laser lines to extraordinary precision.

IV. Conclusion

During the previous contract period, submillimeter detectors have been developed to an advanced stage. The program envisioned for the next three years is expected to bring about a similar advance in the area of sources. This, coupled with sophisticated refinements in integrated detector technology that will be undertaken, will advance the submillimeter state of the art to levels comparable to other regions of the spectrum. New laboboratory physics experiments as well as systems application in a wide variety of fields will become possible.

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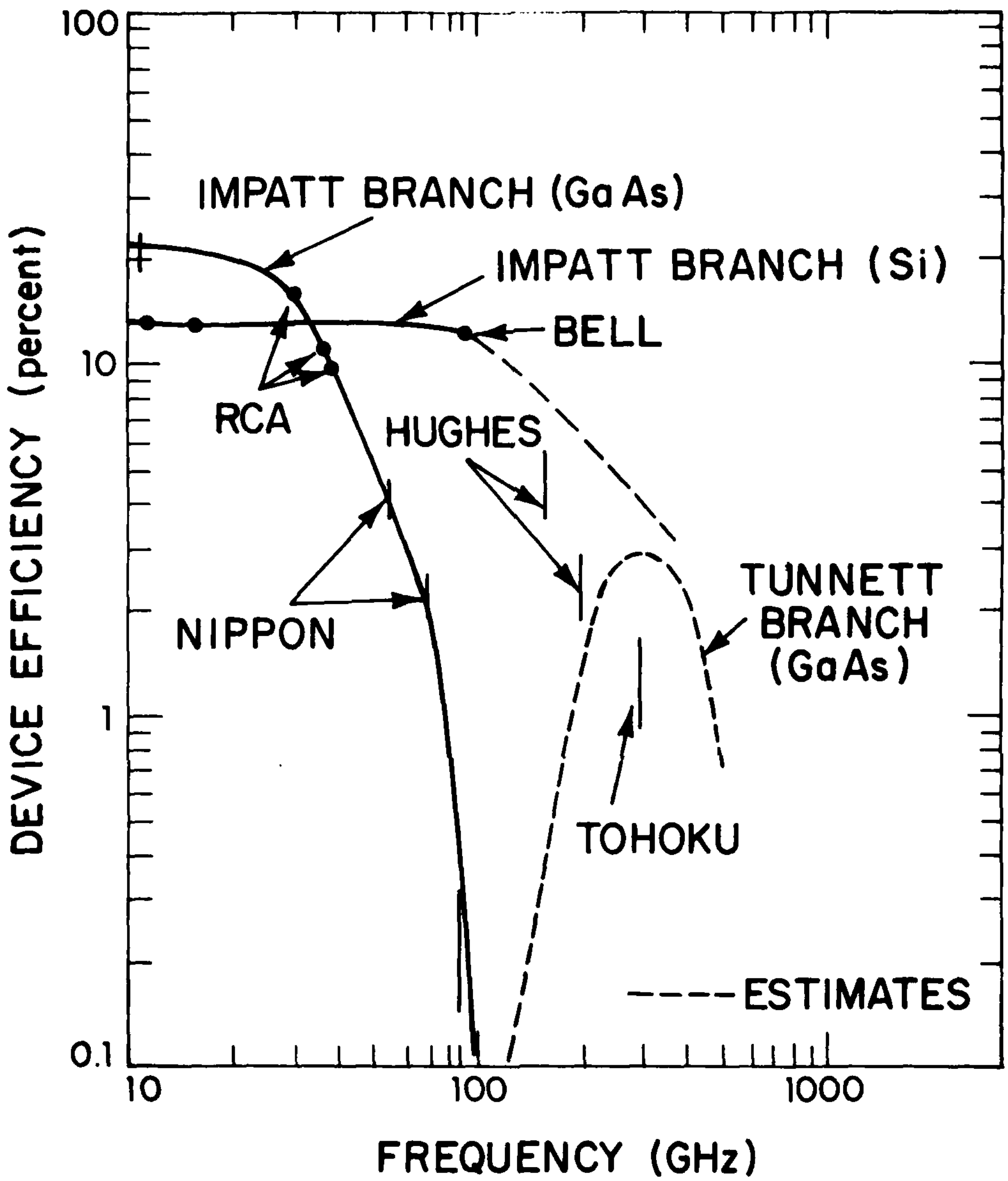

Figure 1

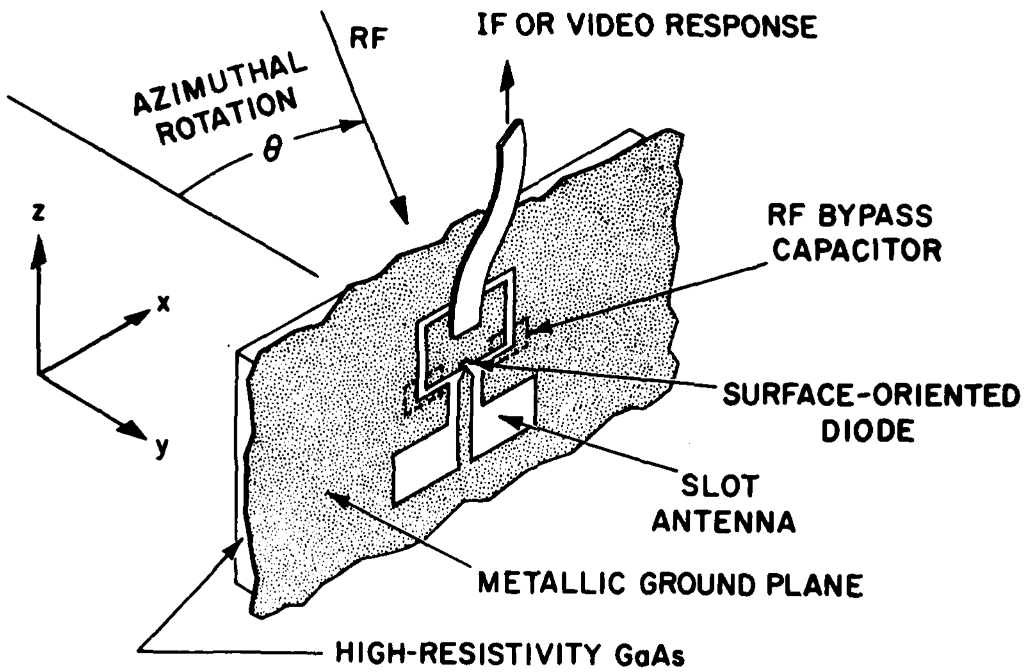

Figure 2a

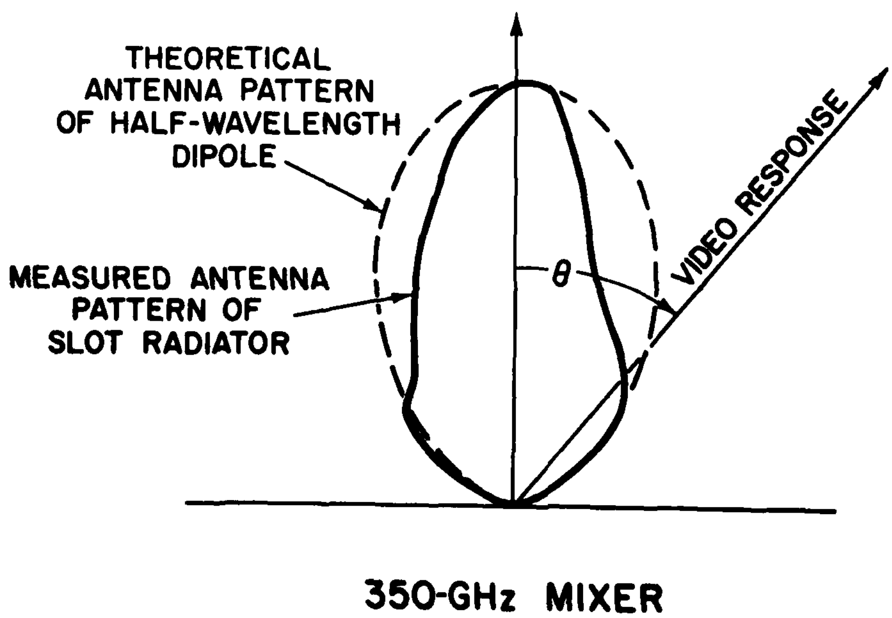

Figure 2b

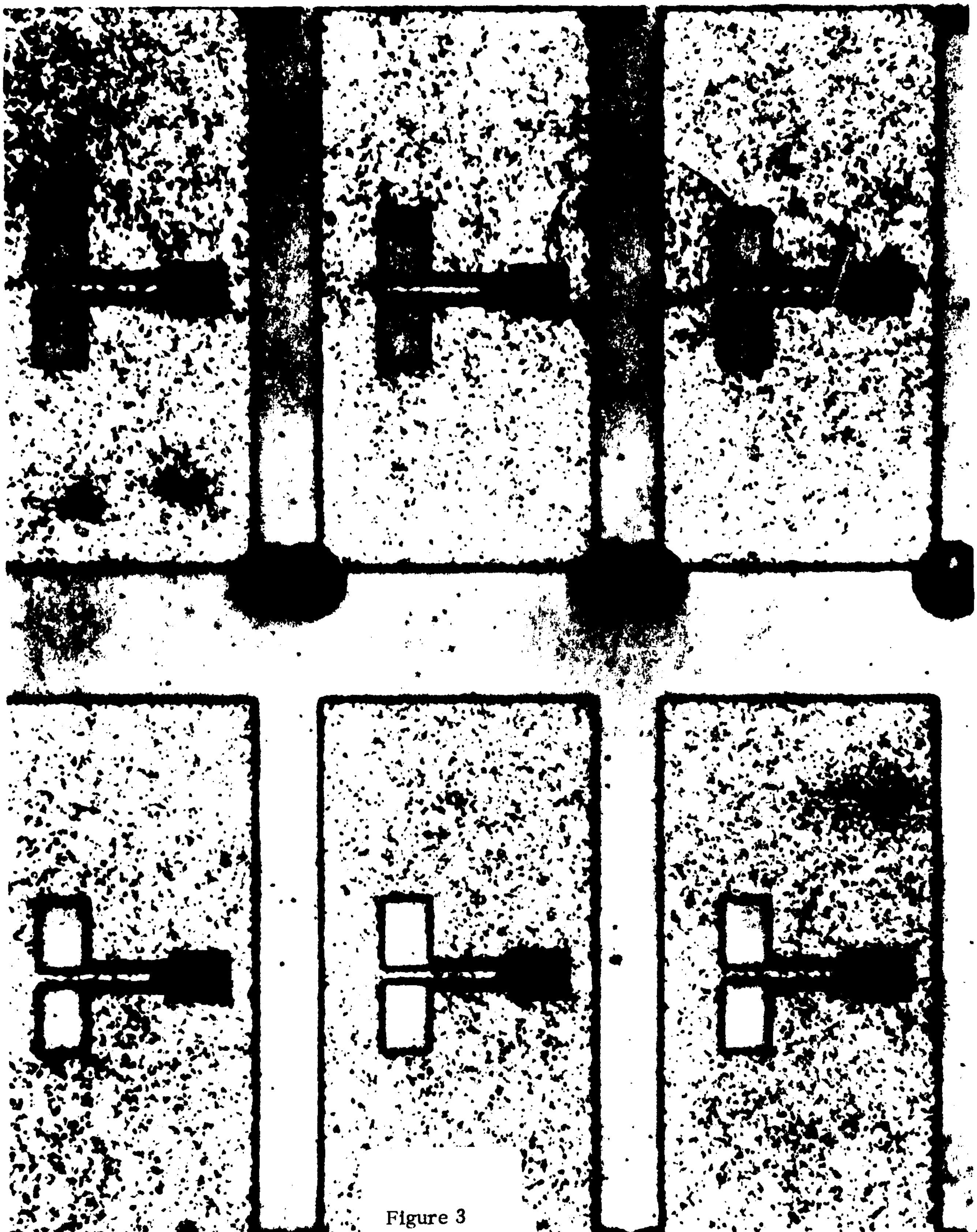

Figure 3

Far-ir heterodyne radiometric measurements with quasioptical Schottky diode mixers[a]

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We have made heterodyne radiometric measurements with GaAs Schottky diode mixers, mounted in a corner-reflector configuration, over the spectral range 170 μm to 1 mm. At 400 μm, system noise temperatures of 9700 K DSB (NEP $= 1.4\times10^{-19}$ W/Hz) and mixer noise temperatures of 5900 K have been achieved. This same quasioptical mixer has also been used to generate 10^{-7} W of tunable radiation suitable for spectroscopic applications.

PACS numbers: 73.30.+y, 42.72.+h, 52.70.Kz, 85.30.Hi

Recent developments in plasma physics, radio astronomy, frequency standards, and satellite-based mapping have created a need for fast sensitive far-infrared detectors. Several groups have been studying the use of GaAs Schottky diode heterodyne receivers using either conventional waveguide[1] or quasioptical approaches.[2] In general, the performance levels that have been achieved at frequencies above 300 GHz have not been comparable with those obtained at millimeter wavelengths. We report here the results of radiometric determinations of noise equivalent powers of a new quasioptical system in the wavelength range of 1 mm to 170 μm. At a frequency of 670 GHz, the highest sensitivity (1.3×10^{-19} W/Hz or 9700 K DSB) to date has been obtained. In addition, this same mixer configuration has been used to produce laser sideband radiation of $\sim10^{-7}$ W tunable over 18 GHz. These results open up new possibilities for far-infrared and submillimeter experiments that require extremely high sensitivities and tunable sources. They demonstrate, that at least up to 1760 GHz, there are no fundamental problems impeding the development of high-sensitivity receivers.

The two key elements of a sensitive high-frequency submillimeter mixer are Schottky diodes having appropriate parameters and a coupling structure that introduces radiation into the diode efficiently. The GaAs

[a] Work was sponsored by the Department of Energy, the National Science Foundation, the U.S. Army Research Office, and the Department of the Air Force.

Schottky diodes used in these experiments were developed especially for high-frequency applications and have been discussed previously.[3] The diodes are typically 1 μm in diameter, have 1.5×10^{-15} F capacitance, and have a series resistance of 45 Ω. A quasioptical approach to the design of a mixer at submillimeter wavelengths was adopted because of the physical difficulties of embedding a diode in a fundamental waveguide at these frequencies. The approach taken, first proposed for the submillimeter by Sauter and Schultz,[4] uses a long, typically four wavelengths, traveling-wave line source set in a corner reflector. The line source is an extended "whisker" point-contacted to the Schottky diode. Impedance and radiation characteristics are determined by the length of the line source, the corner angle, and the separation between line source and corner. Kräutle *et al.*[5] calculated theoretically and demonstrated experimentally the relative magnitude of the gain of a traveling-wave antenna with corner reflector versus antenna alone to be 12 dB. However, in their experimental setup at the time the overall conversion loss was relatively high (29.5 dB).

The radiation pattern of this type of mixer can be analyzed quantitatively by considering a long line source with a traveling-wave excitation. Assuming that the current phase velocity is equal to the free-space phase velocity, the electric field pattern is given by[6] (omitting unnecessary constants)

$$E_\theta(\theta)=\left(\frac{L}{\lambda}\right)\sin\theta\left|\frac{\sin[(\pi L/\lambda)(1-\cos\theta)]}{(\pi L/\lambda)(1-\cos\theta)}\right|,$$

where the length of the line source is given by L/λ and the coordinate system is as shown in Fig. 1(a). Taking the line source in conjunction with a corner reflector having infinitely large conducting planes, the method of images can be used to evaluate the effects of the reflectors for angles of $\alpha=180°/n$, where n is any positive integer.[7] For the 90° corner, with separation between the line source and the vertex of the reflecting planes given by d/λ, the total electric field pattern is given by the superposition of an array of three image elements and the driven element as follows:

$$E(\theta,\Phi)=E_\theta(\theta)A(\theta,\Phi),$$

where $A(\theta,\Phi)=\cos|2\pi(d/\lambda)\sin\theta\cos\Phi|-\cos|2\pi(d/\lambda)\sin\theta \times\sin\Phi|$.

In order to optimize the corner-reflector mixer we have carried out 100 times scale modeling experiments at 6–8 GHz. A design similar to Kräutle *et al*, which looked sufficiently promising for submillimeter tests, was a 90° corner cube (i.e., a 90° corner reflector with a ground plane) with a 4λ antenna length spaced 1.2λ from the corner. The beam pattern obtained from the modeling is shown in Figs. 1(b) and 1(c), together with the theoretically derived patterns. The principal lobe is seen to be at an angle of 25° to the antenna with a roughly circular beam width of 14°×15° (full width at the 3-dB points). Thus the beam shape for the 90° corner reflector is nearly ideal because of the equal beamwidths in the principal planes. Systems now being evaluated with 60° and 45° corner angles have potential for improved beam efficiencies. It should be noted that the 90°, and to a lesser extent the 60° and 45° reflectors, admix the polarizations of the incident radiation fields.

Another essential consideration is for the terminal or radiation impedance of the mount to be matched to the impedance presented by the diode. Model measurements show the receiver's radiation impedance to have a resistive component between 75 and 150 Ω and a reactive component less than 60 Ω over a wide ($\approx 40\%$) bandwidth. Based on our measurements of the diode characteristics, this terminal impedance level presents a substantial mismatch to the mixer diode and offers an important area for future optimization. The signal and local oscillator frequencies are decoupled from the intermediate frequency port by about 13 dB by the bend in the antenna, and no additional filtering is required. This is an inherent advantage of the traveling-wave line source configuration.

The actual corner-reflector diode mount, shown in the inset of Fig. 2, is constructed in two pieces. Each reflector surface has dimensions of 7.9 mm high by 9 mm wide and is highly polished. The ground plane is made from a separate piece of copper and the surface is also highly polished. The 12 5-μm-diam (4λ long at 385 μm) antenna wire is spaced 0.46 mm (1.2λ at 385 μm) from the corner of the reflector and is bonded to a 0.13-mm-diam center conductor which forms part of a 50-Ω coaxial-line i.f. connector. During the diode

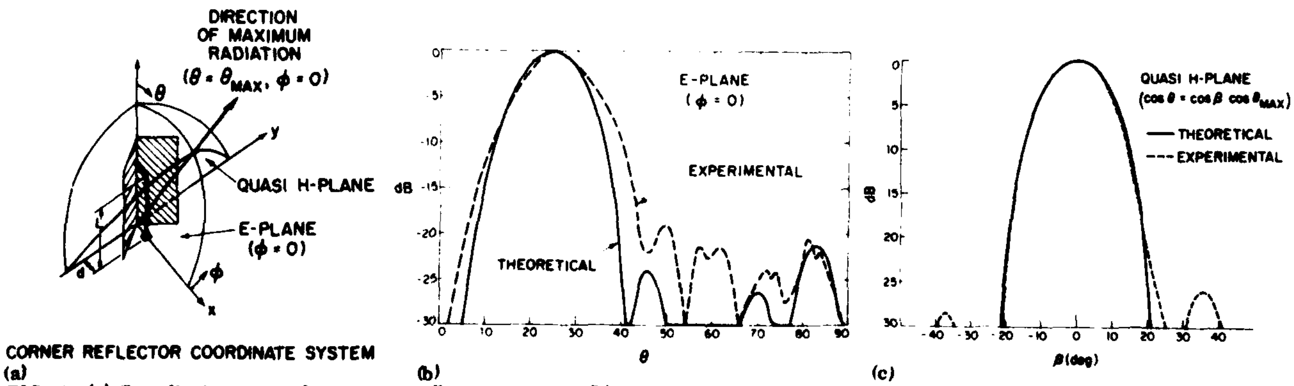

FIG. 1. (a) Coordinate system for corner-reflector antenna. (b) Comparison of theoretical calculations (outlined in text) with experimental data modeled at 7 GHz of corner-reflector antenna pattern about the direction of maximum radiation in the vertical plane. (c) Same for orthogonal plane. β is measured with respect to the direction of maximum radiation.

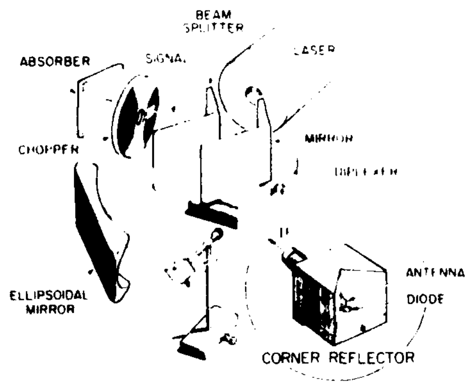

FIG. 2. Quasioptical radiometer operating between 170 μm and 1 mm. Inset shows details of corner-reflector mixer.

contacting operation the chip stud is positioned through a hole in the ground plane, and the etched antenna whisker is lowered into contact with one of the many Schottky barrier diodes on the GaAs chip. A high-power optical microscope is used to observe the diode contacting operation. One advantage of this approach for the corner-reflector mount is the ease with which subsequent diode contacts can be made, if needed. With the corner-reflector mount positioned in the contacting jig, a set screw that locks the diode in position can be released, the chip stud rotated, and a new selected device contacted. Parameter studies and mount optimization can thus be readily and conveniently performed.

We evaluated the performance of the corner-reflector mixer by incorporating it into a Dicke-type radiometer whose components are shown in Fig. 2. Although the mixer was designed along with the rest of the system for approximately 400 μm, several tests were made at both longer and shorter wavelengths to estimate the potential performance. The diplexer, which couples the local oscillator and a corresponding mode from a blackbody into the mixer, consists of a folded interferometer similar in principle to that of NRAO[8] and Erickson.[9] The beam splitters are 3-mil Mylar films stretched over machined reflector mounts, which gave about 50% transmission at the design wavelength. A right-angle ellipsoidal mirror was used to match the signal and LO beams into the antenna pattern of the corner reflector. With this system virtually all of the signal, and better than 90% of the LO, illuminate the mixer. The i.f. amplifier, at 2500 MHz, had a noise temperature of 245 K (2.6 dB) and was operated in series with a 75-MHz bandpass filter. The laser local oscillators consisted of optically pumped far-infrared molecular lasers of standard waveguide design[10] with hybrid output mirrors. They were pumped by a free-running stable CO_2 laser of about 50 W and typically had an output power of about 30 mW. At the operating bias of 1 mA the far-infrared radiation produced a video signal which ranged from 400 to 800 mV. This was found sufficient to saturate the mixer diode at all but the 170-μm line. The identity of each LO line was unambiguously determined by using the diplexer as an interferometer; tuning through approximately 10 interference maxima allowed a wavelength measurement to 0.5% accuracy. A carcinotron was used for the measurements at 1 mm.

The blackbody radiometric measurements were made with RT and liquid-nitrogen-cooled Eccosorb material (AN-72) which alternately illuminated the mixer. Rectified i.f. signals were read directly on a digital voltmeter, and differences in voltage between hot and cold load was synchronously detected in conjunction with the chopper. A standard Y-method[11] interpretation of the data then led to the results shown in Table I for wavelengths near 400 μm, for 1 mm, and 170 μm.

The conversion loss was both calculated from the expression $T_{sys} = T_{mixer} + L_c T_{i.f.}$ and measured directly by changing the temperature of the i.f. input resistor and comparing this with the signal produced by a known ΔT applied to the optical input of the radiometer. In general, the diode noise temperature was near RT and the LO added approximately 100 K excess noise. The diode noise temperatures were measured with an isolator inserted between the diode and the i.f. amplifiers to reduce the effects of changing impedances. Total system noise temperatures were measured without the isolator and are listed in the third column. The best values obtained are given, but variations from diode to diode and mount to mount were typically only about 1000 K.

The results obtained at 1 mm and 170 μm were sufficiently encouraging that quasioptical systems designed specifically for these wavelengths are now being fabricated. Extrapolation from these initial results indicates that this approach can yield usable mixers covering the spectral region from 100 μm to 1 mm with noise temperatures ranging from 3000 to 10 000 K DSB.

Our current results suggest some immediate applications of this technology to far-ir spectroscopy, and we have successfully investigated a number of these. In one such experiment, not described in detail here, we have mixed the pulsed output of a D_2O laser operating at 385 μm (778 870 MHz) with the cw output of a 394-μm (761 607 MHz) formic acid laser in a Schottky diode mixer. The i.f. at 17 GHz demonstrates the wideband capabilities of the mixer; we can thus expect to carry out tunable high-resolution atmospheric spectroscopy using a remote blackbody source (such as the sun) over a broad spectral range about numerous far-ir laser lines.

TABLE I. Results of the standard Y-method interpretation of the data for wavelengths near 400 μm, 1 mm, and 170 μm.

λ (μm)	Frequency (GHz)	Total systems noise temp. (DSB) (K)	Mixer temp. (K)	Conversion loss (dB)
946	316.9	13 000	7800	12.8
447.1	670.5	9700	5900	11.6
432.6	692.9	13 100	6900	11.9
419.6	716.2	13 000	6800	11.9
393.6	761.6	14 500	7600	12.3
170.6	1757.5	370 000[a]	...	...

[a]Diplexer not working at this wavelength.

Still greater resolution and sensitivity for spectroscopy can be obtained in the laboratory by using these mixers as sources of tunable radiation. Radiated sidebands of the laser signal were generated and detected using two corner-reflector diode mixers. The first diode mixer was simultaneously illuminated by a far-infrared laser and fed coaxially by microwave sources ranging from 2.5 to 18 GHz. The second diode mixer was then used as a high sensitivity heterodyne receiver to detect the radiated sidebands of the laser signal. Signals of about 10^{-7} W, tunable over 0.5 cm^{-1}, were observed. Extending this approach further, we investigated the use of these diode mixers as high-order harmonic signal sources. Microwave radiation between 17 and 37 GHz was introduced coaxially onto the diode. Radiation generated as high as the 40th harmonic was then detected by a second diode mixer in our standard heterodyne receiver configuration. In one particular case a 37-GHz GaAs Gunn oscillator coaxially fed the mixer diode, which then generated 761 GHz. This solid-state source coupled with the rugged harmonic mixer provided a compact stable tunable source of about 5×10^{-11} W in the submillimeter. Because of the sensitivity of our heterodyne receiver, the observed S/N exceeded 35 dB on a spectrum analyzer having a 100-kHz bandwidth. High-resolution far-ir spectroscopic studies using both these source-detector systems are now in progress.

In conclusion, we have demonstrated that sensitive detectors systems can be readily extended into the submillimeter and far-infrared regime. These devices are now being introduced into specific applications in plasma diagnostics, radio astronomy, and atmospheric spectroscopy.

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Submillimeter Heterodyne Detection with Planar GaAs Schottky-Barrier Diodes

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Abstract—**Planar surface-oriented Pt/GaAs Schottky-barrier diodes have been fabricated and used to detect signals at submillimeter wavelengths. Video detection has been observed up to frequencies as high as 890 GHz. Harmonic mixing between the ninth harmonic of a 74.21-GHz signal and the second harmonic of 333.95-GHz radiation has also been obtained.**

Introduction

THE coupling of submillimeter-wave or far-infrared laser radiation to Schottky diodes is a formidable problem because of the small device dimensions involved. In the past, carefully formed whiskers have been used to provide contact and to serve as a high-frequency antenna. GaAs Schottky diodes using such whisker structures have been shown to be excellent detectors and mixers for submillimeter radiation [1], [2]. However, further development along these lines requires the physical contacting of even smaller devices having submicron dimensions [3]; this procedure has intrinsic limitations. The objective of the present work is to overcome this difficulty by fabricating small, planar, surface-oriented Schottky diodes, in which both terminals of the rectifying junction lie on the same surface of the GaAs wafer. This single-sided geometry lends itself naturally to an integrated circuit approach with the connection of matched stripline antennas and IF filter networks. In addition, the contacting of small devices becomes more feasible. Although devices with this topography have been fabricated previously [4]–[6], they have heretofore been restricted to frequencies below 100 GHz primarily because their relatively large device areas have led to higher capacitance and lower cutoff frequencies. Diodes with diameters as small as 2 μm have been fabricated in our effort, permitting operation at submillimeter wavelengths for the first time.

Device Construction

The geometry of the device is shown in Fig. 1, together with a scanning electron micrograph of the completed device. To fabricate this structure, two layers of GaAs are epitaxially grown on semi-insulating substrates in a hydrogen transport $AsCl_3$ vapor phase system. The first layer (n^+) is 3 μm thick and has an n-type concentration of 1×10^{18} cm^{-3}. The top layer (n) is 0.5 μm thick and has a concentration of 1×10^{17} cm^{-3}. Sulfur (H_2S) is used to dope both layers. Selective Se^+ ion implantation is then used to decrease the specific resistance of the Au–Ge alloyed ohmic contact. After the formation of the ohmic contact, the diode and ohmic contact areas are shielded by gold, and the wafer is proton bombarded, converting the n- and n^+-layers to high resistivity material in the bombarded regions. The sputter deposited Pt/GaAs Schottky barrier is approximately 2 μm in diameter. Each device is contacted by means of a stripline overlay pattern. A photograph showing an array of these devices on a completed wafer is shown in Fig. 2.

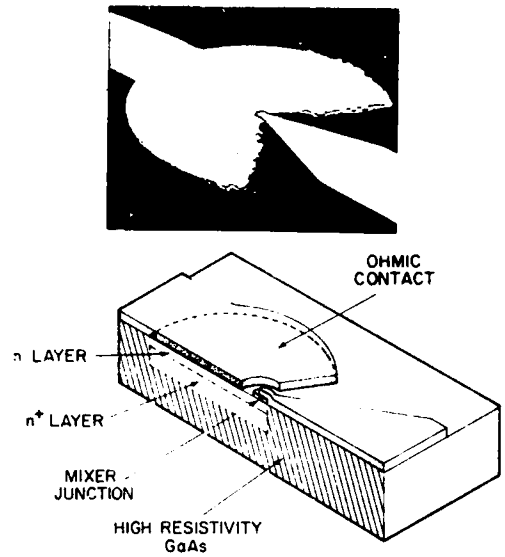

Fig. 1. Bottom: Planar diode as fabricated by growth of n-n^+ epitaxial layers on a high-resistivity GaAs substrate. Top: Scanning electron micrograph showing 2-μm diode (small dot in center), ohmic contact establishing connection to n^+-layer of diode, and two metal strip contacts.

The forward current–voltage (I–V) relationship of a typical device is shown in Fig. 3. This characteristic is quite similar to those of whisker contact Pt/GaAs diodes, with the knee of the nonlinear region occurring at approximately 0.7 V. The n-factors of these devices (describing the deviation from ideal Schottky-barrier characteristics) range from 1.2 to 1.4.

Video Detection

In our preliminary experiments a segment of a device wafer containing a number of diodes was placed on a

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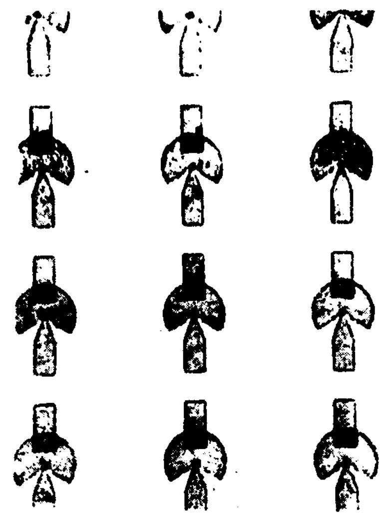

Fig. 2. Array of planar diodes on a GaAs wafer.

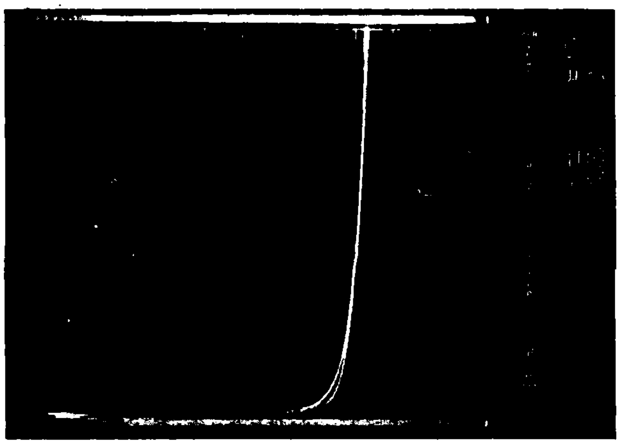

Fig. 3. Forward I-V characteristic of a typical planar diode.

probing apparatus under a microscope so that selected diodes could be contacted while being viewed optically. Various focusing arrangements were used to direct klystron or laser radiation onto the desired spot. In this manner a responsivity of 40 V/W was obtained at 4 mm and about 1 V/W at 337 μm. From these rudimentary results it is not possible to distinguish between intrinsic frequency rolloff of the diodes and variations in coupling inefficiencies at different wavelengths.

Heterodyne Detection

In order to determine the fast response capability of the diodes (that is, demonstrate the nonlinear interaction of high-frequency currents in the device), a number of mixing experiments were carried out. A single device was used which was thermocompression bonded to a stripline on an alumina substrate. First, a carcinotron with an output frequency at 333.95 GHz was mixed with the fifth harmonic of a V-band klystron operating at 66.79 GHz to produce a 90-MHz IF. The S/N obtained was greater than 30 dB. Next, beats were observed, again at a 90-MHz IF, between the ninth harmonic of a 74.21-GHz klystron signal and the second harmonic of the carcinotron operating at 333.95 GHz. Mixing thus occurred at an effective frequency of 668 GHz.

Discussion

Because of their topography, these diodes can be fabricated in large numbers on a single wafer which can be readily integrated with various forms of strip transmission line circuitry. In addition to stripline antennas, numerous other circuit elements can also be integrated with these devices either in a hybrid or monolithic fashion. Some of these concepts have already been successfully demonstrated in the case of thin-film metal-oxide-metal structures [7]. Projected applications of this device involve its use in integrated circuit arrays which are required for large-area coherent detectors and submillimeter imaging devices. Development of simple antennas and arrays is in progress.

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Real-time spectral analysis of far-infrared laser pulses using a SAW dispersive delay line[a)]

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Spectral analysis of high-power pulsed D_2O lasers has been accomplished using SAW dispersive delay lines. Both the contributions of longitudinal modes and the tuning of the stimulated Raman line at 385 μm have been observed.

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We report here a novel technique, using harmonic heterodyne detection in conjunction with a surface acoustic wave (SAW) dispersive delay line, to perform a real-time spectral analysis of laser signals. This has proven to be necessary in the development of pulsed high-power optically pumped submillimeter lasers which are to be used in plasma diagnostics.[1-3] The spectral characterization of these high-power lasers has heretofore been particularly difficult since the repetition period of the 100-nsec pulses is typically from 1 to 10 sec. Also, because of the variability of the pulse behavior from shot to shot, all of the desired frequency information must be obtained on one pulse. This last constraint effectively limits the useful role of high-finesse Fabry-Perot interferometers, which determine only average characteristics. Transient digitizers can, of course, be used in conjunction with a computer capable of performing a near-real-time Fourier transformation. The use of a SAW dispersive delay device provides a simple method of obtaining real-time pulse spectral analysis in this type of experiment. In our initial experiments we have resolved the contribution of three narrow longitudinal modes to a 385-μm D_2O laser pulse spectral linewidth. Furthermore, we have obtained confirmation that the mechanism of lasing is via a stimulated Raman process[4,5] which tunes with a linear dependence on pump frequency.

In pulse measurements that involve frequency analysis, the laser signal must first be heterodyned with a local oscillator to produce an i.f. which is then processed to provide the desired information.. Figure 1 is a diagram of the overall setup. In our experiment we mix the submillimeter laser pulse at 779 GHz (385 μm) with the 11th harmonic of a tunable V-band klystron in a GaAs Schottky diode mixer, producing an i.f. of 420 MHz. The mixer diode was of the planar[6] surface-oriented type used in an open mount. The signal then proceeds to the $LiNbO_3$ dispersive delay line and is displayed on a high-speed storage oscilloscope.

The essential concept for the present application is that frequency components lying within the 100-MHz bandwidth of the SAW device[7] and centered at 420 MHz will be delayed from 4 to 20.3 μsec with a linear dependence on frequency. Since the SAW device produces a total dispersive delay of 16.3 μsec, an oscilloscope display of its output effectively becomes the equivalent to that obtained from a real-time spectrum analyzer. The success of this technique relies strongly on the fact that the signal processing takes place subsequent to external electrical pickup noise. A variable 200-nsec gate further aids in minimizing the electrical noise by allowing signal input to the SAW device only while the 100-nsec laser pulse is on.

The SAW device used as the frequency-dispersive element was originally developed for use as a radar pulse expander and compressor with wide bandwidth and large compression ratio. The basic SAW configuration (also called a

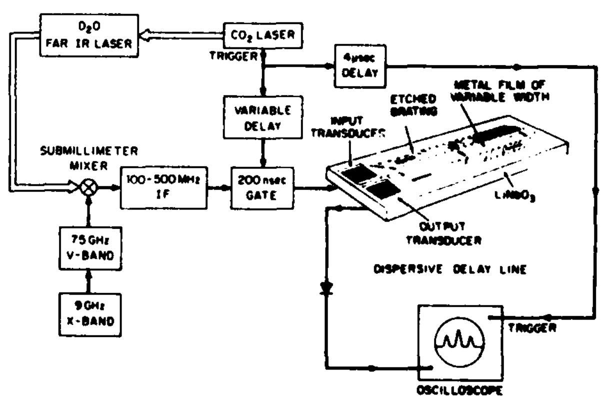

FIG. 1. Experimental setup for pulsed heterodyne experiment. The SAW device is used to perform a real-time spectral analysis of the optically pumped D_2O submillimeter laser signal (the metal film on the SAW device is used to introduce an empirically determined correction to obtain optimum phase characteristics).

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[b)] Supported by the National Science Foundation.

TABLE I. Parameters of reflective array compressor.

Center frequency	420 MHz
Bandwidth Δf	100 MHz
Dispersion T	16.3 μsec
Dispersion-bandwidth product	1630
Constant delay	4 μsec
Material	$LiNbO_3$
Insertion loss	30 dB (flat across passband)

reflective array compressor or RAC) is shown as part of Fig. 1. The input and output interdigital transducers convert an electrical signal to a Rayleigh surface acoustic wave, and vice versa, respectively. The oblique grooves in this device have a spacing that decreases as a function of distance from the input transducer. The surface wave is strongly reflected at a right angle at a point in the reflective array where the groove spacing in the propagation direction matches the wavelength of the surface wave. A second reflection in a symmetrically placed mirror-image grating sends the wave to the output transducer. The groove positions are established such that the surface wave travels from input to output along a path whose length (and group delay) is a linearly increasing function of frequency, a so-called up-chirp. The RAC used in the present experiment was conventional in design: its parameters are given in Table I.

The number of resolvable frequencies is proportional to $(T\Delta f)^{1/2}$, where the proportionality constant is of the order of unity and depends on the precise definition of the resolution. For the present case, the RAC device provides approximately 40 resolution elements or, equivalently, a resolution of about 2.5 MHz. In order to produce a nondeformed spectral image, the requirements $\frac{3}{2}(T\Delta f)^{1/2} < T/\tau < \frac{1}{4}T\Delta f$, where τ is the pulse duration, must be satisfied. For our case $\tau = 100$ nsec , so that $60 < 160 < 400$.

Figure 2 (top) shows the output of the delay line when heterodyning the harmonics of a V-band klystron with a

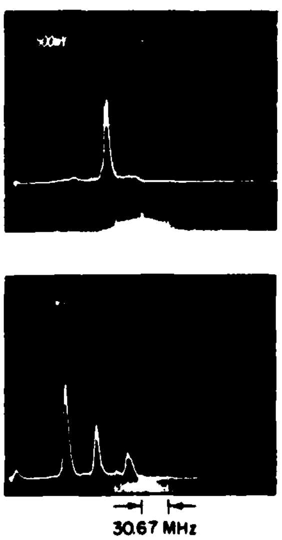

FIG. 2. Oscilloscope display of the output of the SAW dispersive delay line. The upper trace shows, as a function of frequency, the single-mode laser signal at relatively low laser output power. Three longitudinal modes, generated at high laser power, are displayed in the lower trace.

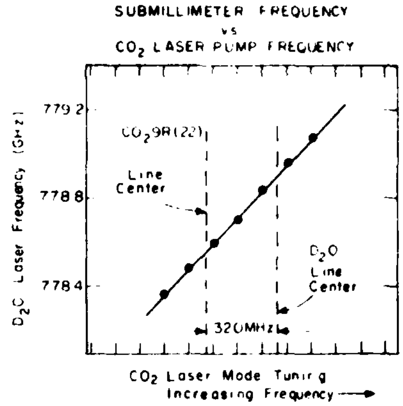

FIG. 3. Submillimeter Raman laser output frequency as a function of the CO_2 laser pump frequency. Each data point was determined by tuning the local oscillator.

D_2O laser pulse from a relatively low-power (~ 50 kW) submillimeter laser oscillator optically pumped with a single-mode CO_2 TEA laser. The submillimeter output is seen to be single mode with a total linewidth of about 7.5 MHz and is quite reproducible. The 30.67-MHz coordinate axis derives from the 16.3-μsec dispersion over a 100-MHz bandwidth. The bottom trace in Fig. 2 shows the same laser operating at higher pump and higher output powers (~ 800 kW). Although every shot is somewhat different in amplitude distribution, we have been able to determine the presence of no more than three narrow adjacent longitudinal modes of the 4-m-long oscillator cavity. The frequency of the V band was determined by mixing with a multiplied X-band stabilized reference to which it can also be locked. This has allowed the V-band to be tuned, the bandwidth of the measurement to be extended, and verification to be obtained that all the modes have been observed. The detection of these narrow modes is a particularly important result since Fabry-Perot studies had indicated that the high-gain D_2O lasing medium may be generating a broad spectral band of superradiance along with more than three longitudinal modes.

At the highest pump levels the 385-μm D_2O laser is expected to be predominately a stimulated Raman process. In this regime of operation the submillimeter output frequency should follow the CO_2 pump frequency. This has been observed in recent measurements[3] using a Fabry-Perot interferometer and tuning the CO_2 with a ZnSe étalon in steps of 112 MHz (the CO_2 oscillator longitudinal mode spacing). Now, using the RAC device and tuning our local oscillator, we have been able to follow, as shown in Fig. 3, the submillimeter output continuously through several hundred MHz. This represents a definitive determination that emission is by a stimulted Raman process and indicates the potential of using this laser system as a source of continuously tunable high-power radiation.

We believe that this is the first time that a SAW dispersive filter has been used to obtain a Fourier transform of a laser pulse for the purpose of studying a laser's spectral characteristics. The output of a dispersive delay line is actually

the Fresnel transform of the input signal. If the time extent of the input signal falls within the limits discussed previously, the Fresnel transform is an adequate approximation of the Fourier transform.[8] This condition holds in the present case; moreover, knowledge of the precise arrival time of the pulse allows exact determination of the frequency-vs-time relation. Since these conditions are satisfied in many pulsed laser experiments, this technique should find wide application in areas such as optical radar signal processing as well as in the spectral analysis of fast time-dependent laser-related phenomena. Furthermore, we foresee that an extension of this type of signal processing, the chirp transform,[9] will also have an important role in more sophisticated spectral analysis of fast time-dependent phenomena.

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EXCITED STATE SPECTROSCOPY AND KINETICS OF CH_3F USING TUNABLE SUBMILLIMETER SOURCES*

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Abstract

The generation and detection of tunable submillimeter radiation has been demonstrated using optically pumped lasers and quasi-optical Schottky diode mixers and detectors. We have now used these tunable sources to investigate the collision induced infrared-submillimeter double resonance spectra of CH_3F. In these experiments the ν_3 vibrational state of CH_3F is populated using either cw or pulsed CO_2 laser radiation. Tunable submillimeter radiation, generated as a laser sideband, is then used to probe the rotational levels in the excited state. This double resonance technique is applicable to a wide class of molecules, and is particularly important for those systems which are candidates for optically pumped submillimeter lasers.

The system using cw laser pump excitation is primarily suited to obtaining spectroscopic constants with high precision. For example, the low lying levels of the ν_3 state of CH_3F were populated by collisions after driving the $^qQ(12,1)$ and $^qQ(12,2)$ rovibronic transitions with the 9P(20) line of a cw CO_2 laser. The $J=7\rightarrow 8$, $\Delta K=0$, $K=0,\ldots 7$ and $J=12\rightarrow 13$, $\Delta K=0$, $K=0,\ldots 4$ series of rotational transitions in the ν_3 state were then observed. The best fit to the line centers of the $J=7\rightarrow 8$ and $J=12\rightarrow 13$ series of transitions provided improved values of the centrifugal distortion constants $D_{J1}=0.0568\pm 0.0005$ MHz and $D_{JK1}=0.517\pm 0.004$ MHz.

The submillimeter heterodyne receiver system was fast enough to allow the observation of rapid radiative and collisional relaxation processes among rotational levels. The CH_3F was pumped to ν_3, $J=12$ states with 200 ns pulses from a CO_2 TEA laser. The kinetics of the excited state population as well as stimulated emission and superfluorescence were observed for various rotational transitions in the ν_3 state. With these techniques it is now possible to investigate directly the rotational spectra and kinetics of collision induced population transfer among rotational levels of an excited state. Thus, this infrared-submillimeter double resonance technique will have general applicability to any molecule which can be optically pumped.

See the following 3 figures

*This work was sponsored by the Army Research Office and the Department of the Air Force.

Figure Captions

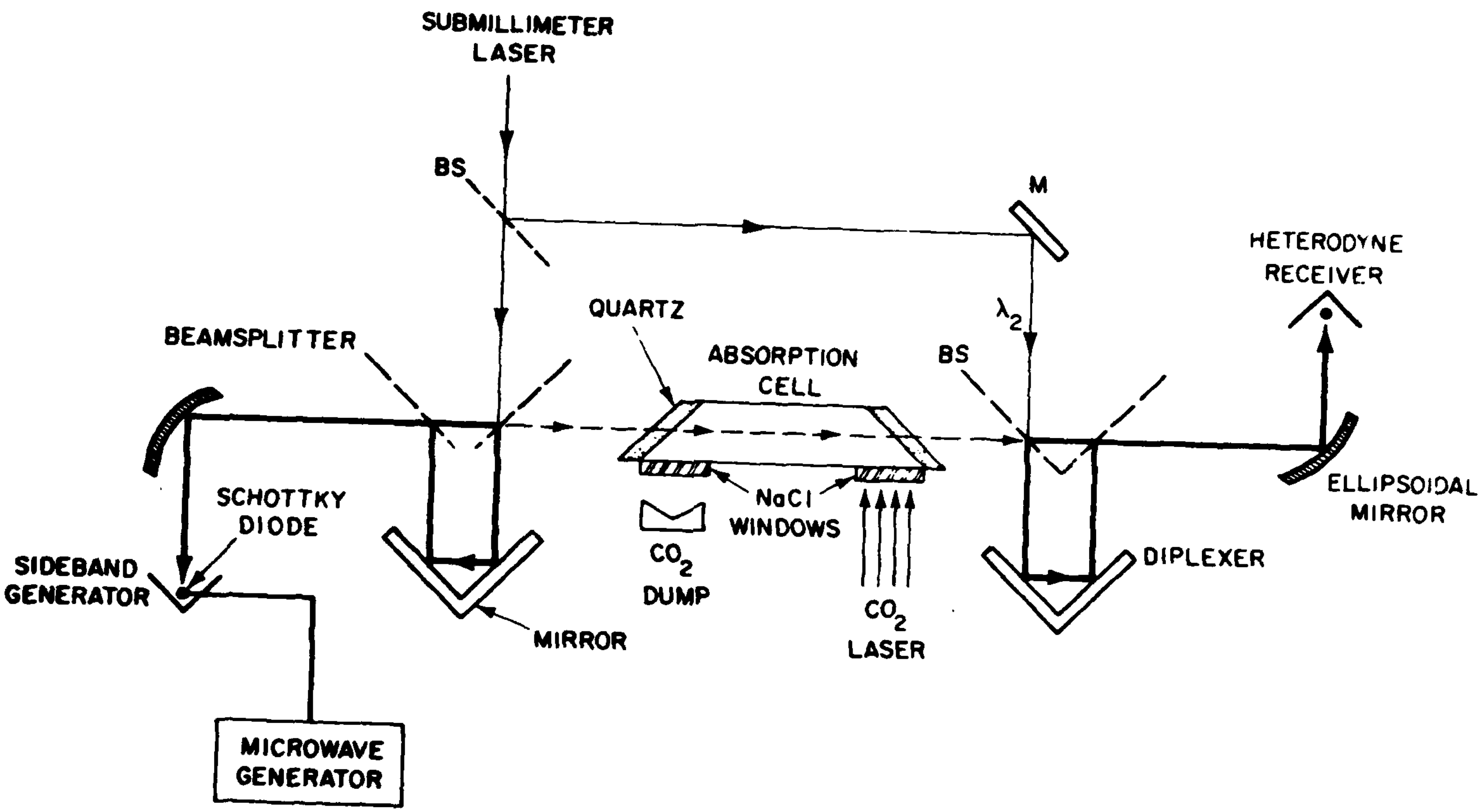
Figure 1. Schematic diagram of the double resonance spectrometer. The intensity of the sideband beams transmitted through the sample cell was 40 dB above the receiver noise level.

Figure 2. Schematic energy level diagram of CH_3F showing the infrared vibrational transitions (dark arrows) pumped by the 9P(20) CO_2 laser line and the submillimeter rotational transitions (light arrows) observed in the cw double resonance experiment. J is the total rotational angular momentum and K is the component of J along the C-F axis.

Figure 3. (a) Stimulated emission pulse followed by absorption of the CH_3F submillimeter laser beam at a pressure of 25 mTorr.

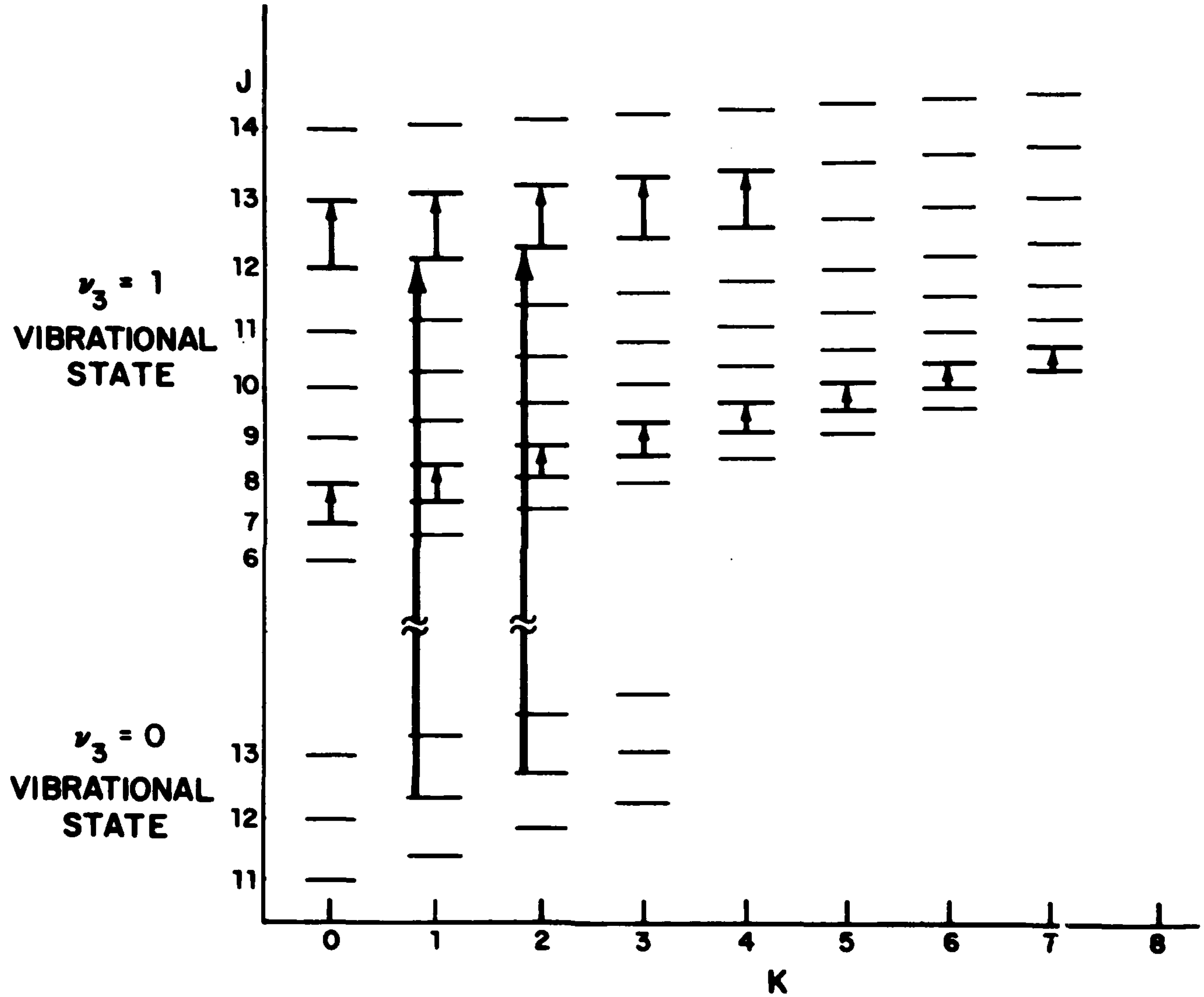
(b) Absorption of a sideband beam tuned to the $(\nu_3, J, K) = (1, 7, 2) \rightarrow (1, 8, 2)$ transition at a pressure of 1.19 Torr.

(c) Superfluorescence pulse followed by absorption of a sideband beam tuned to the $(\nu_3, J, K) = (1, 7, 2) \rightarrow (1, 8, 2)$ transition at a pressure of 1.19 Torr.



DOUBLE RESONANCE SPECTROMETER

Figure A-1



J
14
13
12
11
10
9
8
7
6
ν_3 = 1
VIBRATIONAL
STATE
ν_3 = 0
VIBRATIONAL
STATE
13
12
11
0
1
2
3
4
5
6
7
8
K

Figure A-2

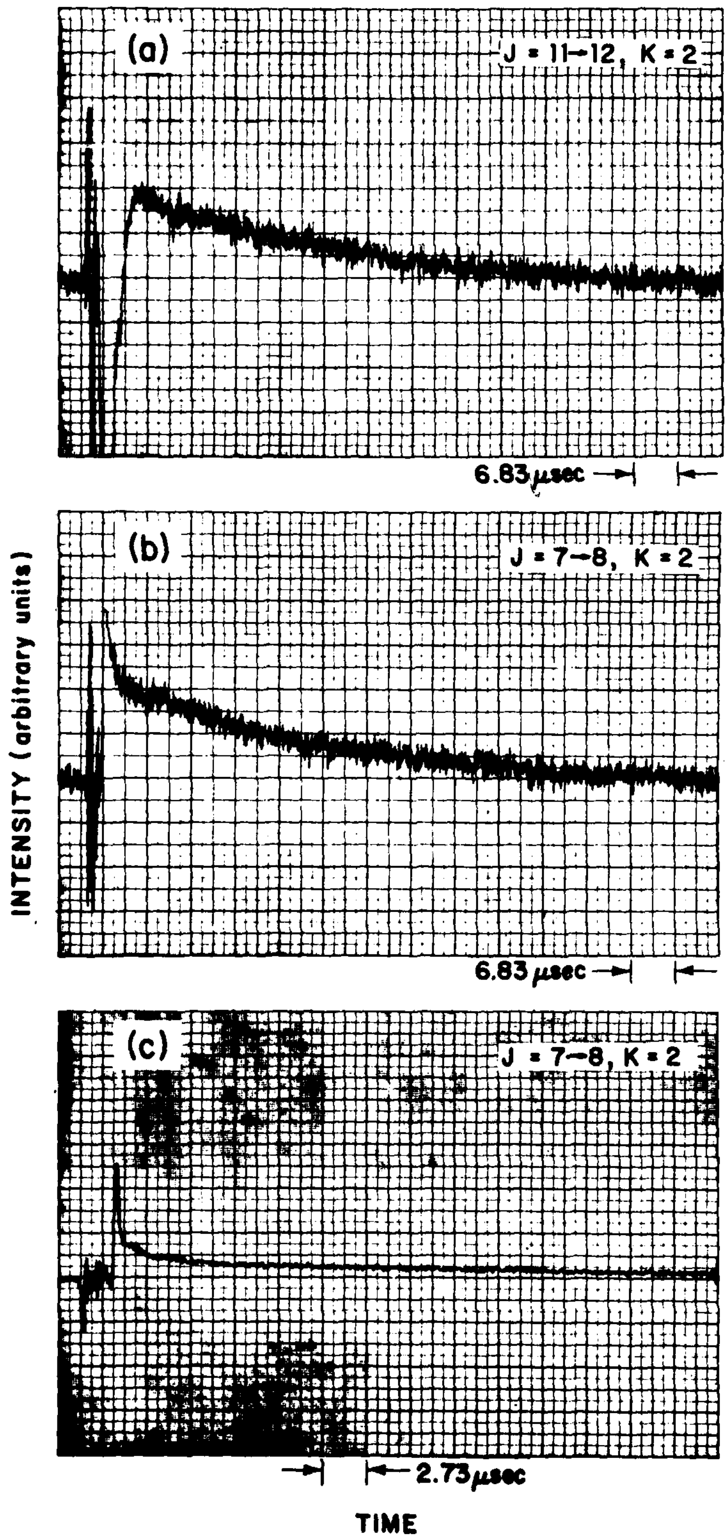

(a)
J = 11→12, K = 2
6.83 μsec
(b)
J = 7→8, K = 2
6.83 μsec
(c)
J = 7→8, K = 2
2.73 μsec
INTENSITY (arbitrary units)
TIME

Figure A-3

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MILLIMETER AND SUBMILLIMETER WAVE PROPAGATION AND CIRCUITS

NORTH ATLANTIC TREATY ORGANIZATION

ADVANCES IN GaAs SCHOTTKY DIODE SUBMILLIMETER HETERODYNE RECEIVERS AND RADIOMETERS*

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SUMMARY

Radiometric sensitivity measurements have been made on a quasi-optical receiver in the spectral range 170 μm to 1 mm. Using GaAs Schottky mixer diodes in a corner reflector configuration, a total system noise temperature of 9,700 K (DSB), or an NEP of 1.3×10^{-19}W/Hz, has been obtained at 447 μm. This same quasi-optical mixer has also been used for the generation of tunable harmonic and side-band radiation suitable for submillimeter spectroscopic applications. Planar, surface-oriented GaAs Schottky diodes have been fabricated by means of photolithographic techniques in conjunction with ion implantation and proton bombardment. High-order harmonic mixing and direct heterodyne mixing with lasers up to 761 GHz have been achieved. These planar diodes can be fabricated into array configurations by means of an integrated circuit approach.

1. INTRODUCTION

Recent developments in radio astronomy, plasma physics, frequency standards and spectroscopy, atmospheric propagation, aeronomy, and in high-altitude aircraft and satellite-based surveillance have created a need for fast, sensitive far infrared detectors. Several groups have been studying the use of GaAs Schottky diode heterodyne receivers using either conventional waveguide (MC COLL, M., 1977) or quasi-optical approaches (GUISTINCIC, J. J., 1977). In general, the performance levels that have been achieved at frequencies above 300 GHz have not been comparable with those obtained at millimeter wavelengths. We report here the results of radiometric determinations of noise equivalent powers of a new quasi-optical system in the wavelength range of 1 mm to 170 μm. At a frequency of 671 GHz (447 μm) the highest sensitivity reported to date has been obtained (FETTERMAN, H. R., 1977)--namely, an NEP of 1.3×10^{-19}W/Hz, or an equivalent system noise temperature of 9,700°K (DSB). The quasi-optical Schottky diode mixer that was used in the heterodyne receiver has also been used to produce far IR laser sideband radiation of $\sim 10^{-7}$ watts tunable over 18 GHz, and to harmonically generate tunable submillimeter radiation up to 760 GHz. Application of this tunable source to spectroscopic problems has been demonstrated. These developments open up new possibilities for far infrared and submillimeter experiments which require extremely high sensitivities.

During the past year we have also developed the next generation of submillimeter detectors--planar, surface-oriented GaAs Schottky diodes fabricated by photolithographic techniques in conjunction with ion implantation and proton bombardment. In contrast to point-contact whisker diodes, here both terminals of the rectifying junction lie on the same surface of the GaAs wafer. This single-sided geometry lends itself naturally to an integrated circuit approach with the connection of matched stripline antennas and IF filter networks. These planar diodes have been successfully used as heterodyne mixers up to 779 GHz, including high-resolution detection of pulsed far IR radiation.

2. RADIOMETER/RECEIVER SYSTEM

The experimental system used in these measurements is essentially a Dicke type radiometer which mixes a laser local oscillator (LO) with radiation from a black body whose radiated power is solely a function of its temperature. Illustrated in Fig. 1, the system consists primarily of the following components: 1) corner-reflector diode mount, 2) ellipsoidal coupling mirror, 3) a quasi-optical diplexer, 4) laser local oscillator and 5) low temperature absorber. The GaAs Schottky diode mounted in the corner reflector was developed especially for high frequency applications and has been discussed elsewhere (CLIFTON, B. J., 1977). The diodes are typically 1 μm in diameter, have 1.5×10^{-15}F capacitance and a series resistance of 45 Ω.

A quasi-optical approach for the mixer was adopted because of the physical difficulties of embedding a diode in a fundamental waveguide at these frequencies. The approach taken, first proposed for the submillimeter by Sauter and Schultz (SAUTER, E., 1977), uses a long--typically four wavelengths--traveling wave-line source set in a corner reflector. The line source is an extended "whisker" point-contacted to the Schottky diode. Impedance and radiation characterisitics are determined by the length of the line source, the corner angle, and the separation between line source and corner.

In order to optimize a corner reflector mount we have carried out 100 times scale modeling experiments at 6 to 8 GHz. One design, similar to Kräutle (KRÄUTLE, H., 1977), which looked promising for actual submillimeter construction, is a 90° corner cube (i.e., 90° corner reflector with a ground plane) with a 4 λ antenna length spaced 1.2 λ from the corner. The beam pattern obtained from the modeling is shown in Fig. 2. The principal lobe is seen to be at an angle of 25° to the antenna, and has a roughly circular cross-section of beam width 14° x 15° (full width at 3 dB points).

The local oscillator and a corresponding mode from the black body (Eccosorb AN-72) at liquid nitrogen temperature were coupled to the mixer utilizing a diplexer. This diplexer was essentially a folded, double interferometer, similar in principle to that used by NRAO (GOLDSMITH, P. F., 1977) and Erickson (ERICKSON, N. R., 1977), but made specifically for the 0.4 or 0.9 mm wavelength region. The beam splitters are 3 mil mylar stretched over machined reflector mounts and gave roughly 50% transmission. An extremely short focal length, right angle, ellipsoidal mirror was used to match the transmitted signal and LO to the antenna pattern of the diode. With this system virtually all of the signal and better than 90% of the LO were transmitted to the detector.

The local oscillators consisted of optically pumped far infrared lasers for the submillimeter and a carcinotron for 1 mm. The laser LO was pumped by a free-running, stable CO_2 laser of about 50 W and had an output power of about 30 mW. This was found sufficient to saturate the mixer diode at all but the 170 μm line. The identity of each LO line was unambiguously determined by using the diplexer as an interferometer; tuning through approximately 10 interference maxima allowed a wavelength measurement to 0.5% accuracy.

*This work was supported by the Department of the Air Force, the U.S. Army Research Office, the National Science Foundation, and the Department of Energy.

3. RADIOMETRIC MEASUREMENTS

The results of our measurements are summarized in Table 1. Two separate corner reflectors were used to cover the wavelengths listed - one designed for 400 μm and a second one for 0.9 mm. For 170 μm neither the diode mount nor the diplexer was optimized, but a result is quoted to show that a black body heterodyne measurement was possible. The rectified IF signal was read on a digital voltmeter, and the difference in voltages between the hot (room temperature absorber or chopper) and cold absorber was synchronously detected. The amplifier had a measured noise temperature of 245 K (2.6 dB). A standard Y-method interpretation of the data then yielded the results shown in the table.

The conversion loss was both calculated from the expression $T_{sys} = T_{mixer} + L_c T_{IF}$ and measured directly by changing the temperature of the IF input resistor and comparing this with the signal produced by a known ΔT applied to the optical input of the radiometer. In general the diode noise temperature was near room temperature, and the LO added approximately 100 K excess noise. The diode noise temperatures were measured with an isolator inserted between the diode and the IF amplifiers to reduce the effects of changing impedances. Total system noise temperatures were measured without the isolator and are listed in the third column. The best values obtained are given, but variations from diode to diode and mount to mount were typically only about 1000 K.

Table 1

λ (μm)	Frequency (GHz)	Total Systems Noise Temp. (DSB) (K)	Mixer Temp. (K)	Conversion Loss (dB)
887.0	338.2	8,200	4,000	9.3
447.1	670.5	9,700	5,900	11.6
432.6	692.9	13,100	6,900	11.9
419.6	716.2	13,000	6,800	11.9
393.6	761.6	14,500	7,600	12.3
170.6	1757.5	370,000*	--	--

*Diplexer not working at this wavelength

4. PLANAR, SURFACE-ORIENTED DIODES

Although the coupling of submillimeter radiation to a single Schottky mixer diode has been successfully solved with the quasi-optical approach of using a corner reflector, the discrete nature of the whisker contacting the diode chip precludes fabrication of arrays of detectors integrated directly into circuits. The objective of this phase of the work is to overcome this limitation by fabricating small, planar, surface-oriented Schottky diodes in which both terminals of the rectifying junction lie on the same surface of the GaAs wafer. This single-sided geometry lends itself naturally to an integrated circuit approach with the connection of matched stripline antennas and IF filter networks. In addition, the contacting of small devices becomes more feasible. Although devices with this topography have been fabricated previously (ALLEN, R. P. G., 1973; BALLAMY, W. C., 1976; WOOD, E. J., 1976), they have heretofore been restricted to frequencies below 100 GHz primarily because their relatively large device areas have led to higher capacitance and lower cutoff frequencies. Diodes with diameters as small as 2 μm have been fabricated in our effort, permitting operation at submillimeter wavelengths for the first time (MURPHY, R. A., 1977).

4.1. Device Construction

The geometry of the device is shown in Fig. 3. To fabricate this structure, two layers of GaAs are epitaxially grown on semi-insulating substrates in a hydrogen transport $AsCl_3$ vapor phase system. The first layer (n^+) is 3 μm thick and has an n-type concentration of $1 \times 10^{18} cm^{-3}$. The top layer (n) is 0.2 μm thick and has a concentration of $1\text{-}2 \times 10^{17} cm^{-3}$. Sulfur ($H_2S$) is used to dope both layers. Selective Se^+ ion implantation is then used to decrease the specific resistance of the Au-Ge alloyed ohmic contact. After the formation of the ohmic contact, the diode and ohmic contact areas are shielded by gold, and the wafer is proton bombarded, converting the n- and n^+-layers to high resistivity material in the bombarded regions. The e-beam deposited Ti/GaAs Schottky barrier is approximately 2 μm in diameter. Each device is contacted by means of a stripline overlay pattern. A photograph showing an array of these devices on a completed wafer is shown in Fig. 4.

The forward current-voltage (I-V) characteristic of a typical device is quite similar to those of whisker contact Pt/GaAs diodes, with the knee of the nonlinear region occurring at approximately 0.7V. The n-factors of these devices (describing the deviation from ideal Schottky-barrier behavior) range from 1.2 to 1.5.

4.2. Submillimeter Measurements

Figure 5 shows a device to which a planar radiating structure has been attached. The metal pattern was designed as a half-wavelength dipole at around 0.9 mm wavelength. The measured radiation pattern shows complex lobe structures apparently because of interference from other radiating elements. A typical radiation pattern obtained at 348 GHz is shown in Fig. 6. Our measurements indicate that modes are excited in the GaAs substrate, with the result that the whole GaAs slab acted as a radiating element. Even though some of the lobes are quite narrow, it is desirable to work with simpler and more controllable structures. Consequently we have begun to make measurements upon scale models of slot and coplanar antennas. One such design which has been modeled is shown in Fig. 7. The measurements have indicated that energy can be coupled quite efficiently to the nonlinear junction with these radiating structures. Submillimeter devices using such structures will be fabricated. Other antenna configurations are also under consideration.

Although no accurate quantitative measurements of the sensitivity of these devices have been made, they have been successfully used as detectors and mixers well into the submillimeter wavelength regime. It has become apparent in heterodyning with pulsed laser sources that in electrically noisy environments planar diodes show better survival and noise suppression characterisitics than conventional whisker-contacted diodes and in addition display remarkable physical ruggedness. High-order harmonic mixing has been obtained by coupling a 9.2879 GHz signal, introduced into the IF line, and a 862.196 GHz (393.6 μm) laser line of formic acid to a planar device. The IF response corresponded to

mixing of the 82nd harmonic of the X-band signal with the laser and had a signal-to-noise ratio of better than 35 dB. This same system of harmonic mixing has been used to phase-lock our 1 mm carcinotron and is used routinely for that purpose.

4.3. Diode and Antenna Arrays

Because of their topography, these diodes can be fabricated in large numbers on a single wafer and thus provide the unique possibilities of array detection and submillimeter imaging. We plan to develop planar imaging arrays composed of planar antennas, surface-oriented mixer diodes, and low-noise FET amplifiers, all integrated on a single chip. Beam steering is also possible using phased-array techniques.

5. APPLICATIONS TO TUNABLE SUBMILLIMETER SPECTROSCOPY

5.1. Spectroscopy with a Black Body

Our current results suggest some immediate applications of this technology to far IR spectroscopy, and we have investigated a number of these. In one such experiment, not described in detail here, we have mixed the pulsed output of a D_2O laser operating at 385 μm (778, 870 MHz) with the cw output of a 394 μm (791, 607 MHz) formic acid laser in a Schottky diode mixer. The IF was at 17 GHz. In another experiment we have mixed two rotational cw lines of a formic acid laser at 393.6 and 405.6 μm; the resultant IF at 22.45 GHz was heterodyned again with the second harmonic of an X-band source fed into the IF port of the diode. These experiments demonstrate the wide-band capabilities of the mixer. We can thus expect to carry out tunable high-resolution spectroscopic measurements against a black body in the laboratory, as well as in a number of terrestrial and space applications using the sun as a remote source.

5.2. Generation of Tunable Far Infrared Radiation

Concurrent with the heterodyne detector effort in the far infrared/submillimeter region we are seeking sources of tunable radiation for applications to spectroscopy, to frequency standards and to remote sensing. The GaAs Schottky diode mixer which we have developed for our sensitive receiver and radiometer can be used as a source of tunable laser-sideband radiation and high-order microwave harmonics. Since the corner reflector configuration acts as an efficient receiver of radiation, it will act reciprocally as an efficient radiator. The resistive nonlinearity of the diode provides the mixing and harmonic generation mechanism.

In our initial experiments, radiated sidebands of a CH_3I submillimeter laser line at 447 μm were generated and detected using two corner-reflector diode mixers. One diode mixer was simultaneously illuminated by the far infrared laser and fed coaxially by microwave sources ranging from 2.5 to 18 GHz. A second diode mixer was then used as a high sensitivity heterodyne receiver to detect the radiated sidebands of the laser signal. Signals of about 10^{-7} watts, continuously tunable over 0.5 cm^{-1}, were observed. Extending this approach further, we investigated the use of these diode mixers as high-order harmonic signal generators. The experimental arrangement is shown in Fig. 8, with the microwave radiation between 17 and 37 GHz introduced via the IF connector line to the diode. Radiation generated as high as the 40th harmonic was then detected by a second diode mixer in our standard heterodyne receiver configuration. In one particular case, a 37 GHz GaAs Gunn oscillator was used to generate 761 GHz. This solid state source coupled with the rugged harmonic mixer provided a compact, stable, tunable source of about 5×10^{-11}W in the submillimeter. Because of the sensitivity of our heterodyne receiver, the observed S/N exceeded 35 dB on a spectrum analyzer having a 100 kHz bandwidth.

High resolution far infrared spectroscopic studies using both these source-detector systems are now underway. As a first demonstration, an absorption cell containing D_2O gas at low pressure was placed in one arm of the spectrometer, as shown in Fig. 8. 35.7 GHz radiation from a microwave source was used to drive the mixer diode, and the 17th harmonic generated (607.345 GHz), after passing through the absorption cell, was detected by the heterodyne receiver. The LO was supplied by an optically-pumped CH_3F laser operating at 604.295 GHz, giving an IF around 3 GHz. Tuning through the D_2O absorption line was accomplished by tuning the microwave source. Figure 9 shows the far infrared output with and without the gas. Because of residual pressure from condensed liquid in the absorption cell, the linewidth could not be reduced below the 20 MHz shown. The resolution of the spectrometer is about 100 kHz, being presently limited by the stability of the laser local oscillator. The design resolution of this system is 10 kHz (3×10^{-7} cm^{-1}) with continuous tunability of ≈ 1 cm^{-1}.

This preliminary work will now be continued with other gases that have absorptions near strong laser LO lines, and will be applied particularly to atomic fine structure resonance lines that have extremely narrow and stable transitions which can be used as references for far infrared frequency standards.

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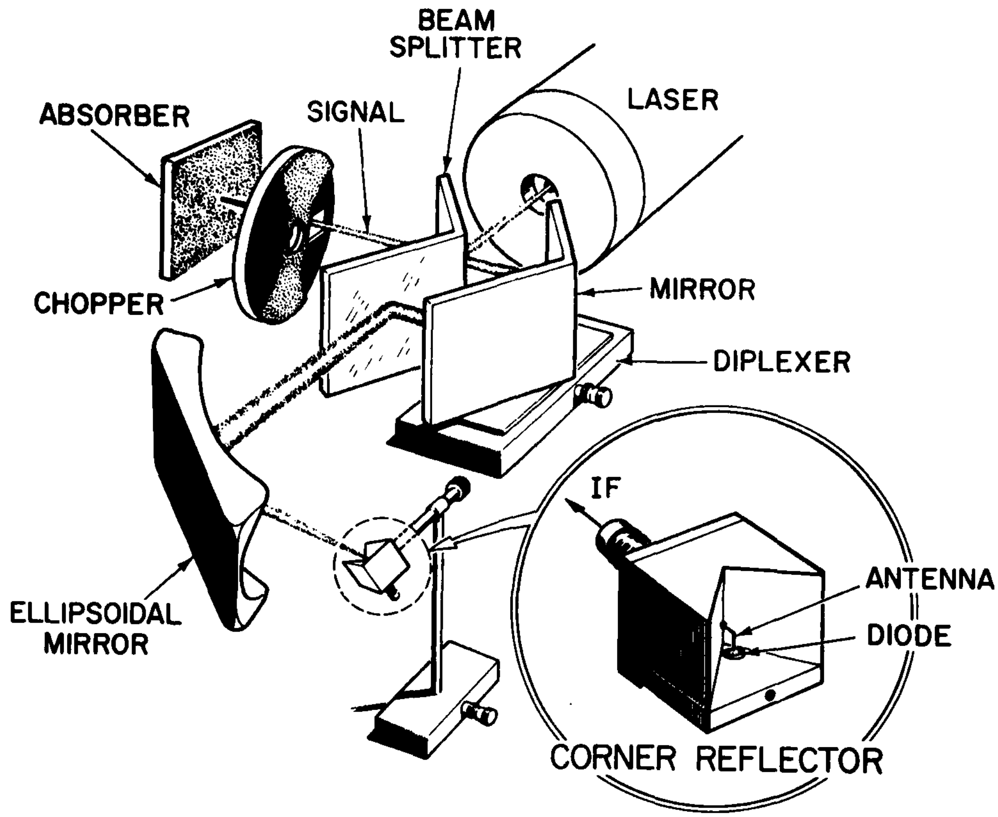

Fig. 1 Quasi-optical submillimeter receiver/radiometer operating between 170 μm and 1 mm.

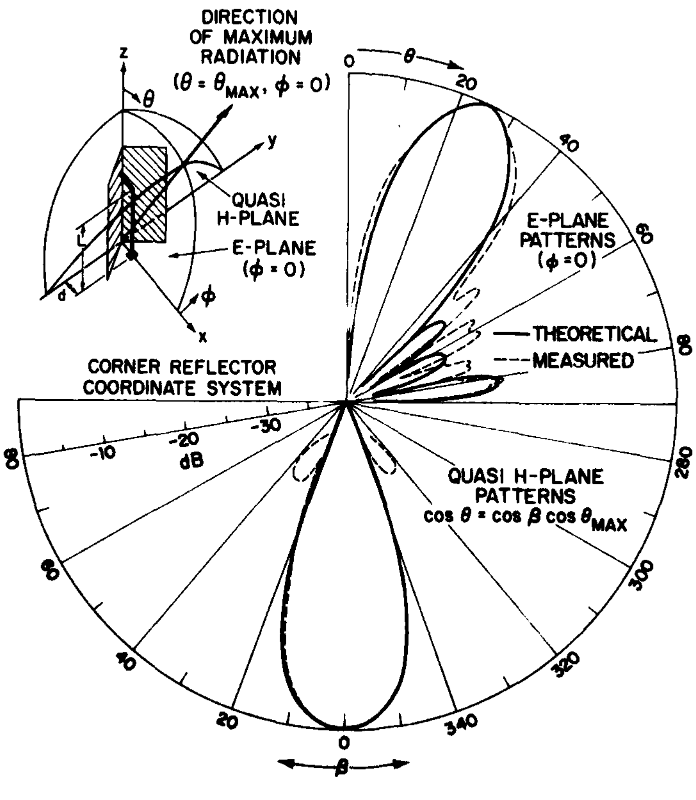

Fig. 2 Comparison of theoretical calculations with experimental data modeled at 7 GHz of corner-reflector antenna pattern.

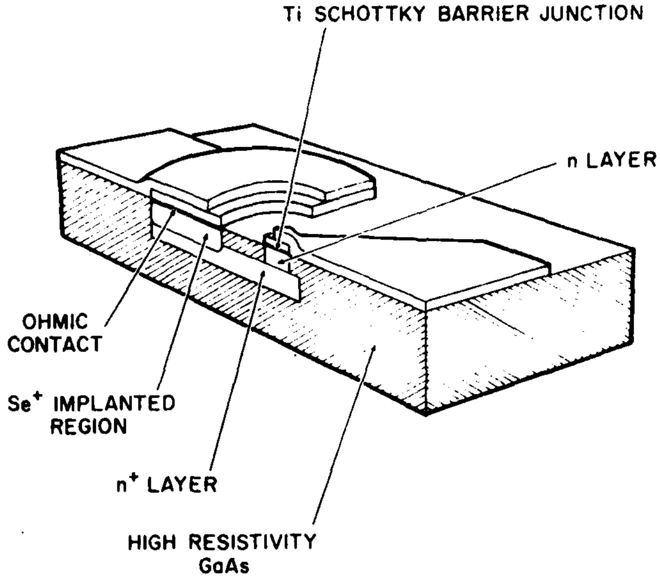

Fig. 3 Planar, surface-oriented diode as fabricated by growth of $n\text{-}n^{+}$ epitaxial layers on high-resistivity GaAs substrate.

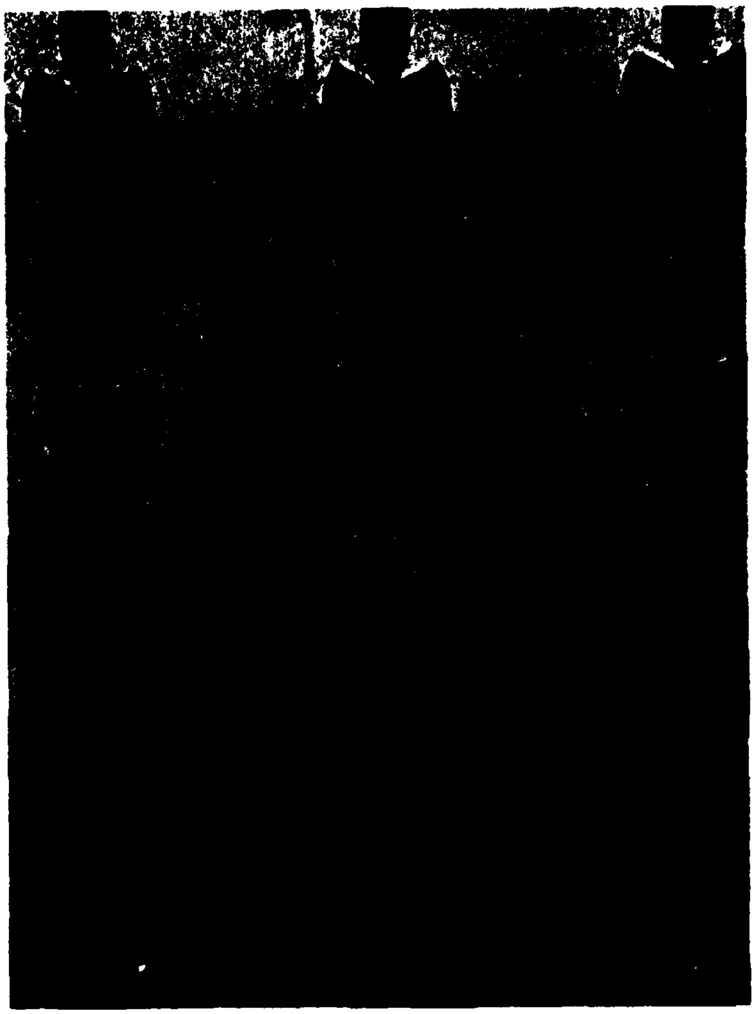

Fig. 4 An array of planar diodes on a GaAs wafer.

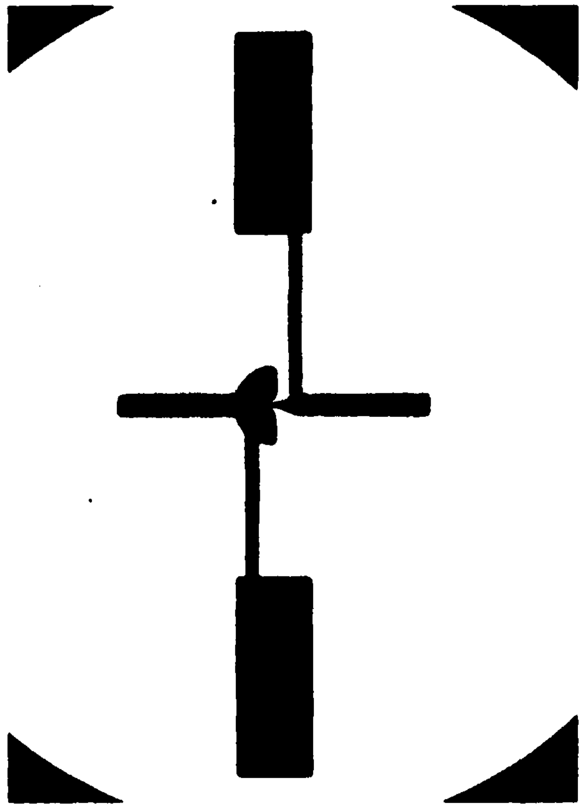

Fig. 5 Planar diode with attached half-wavelength dipole.

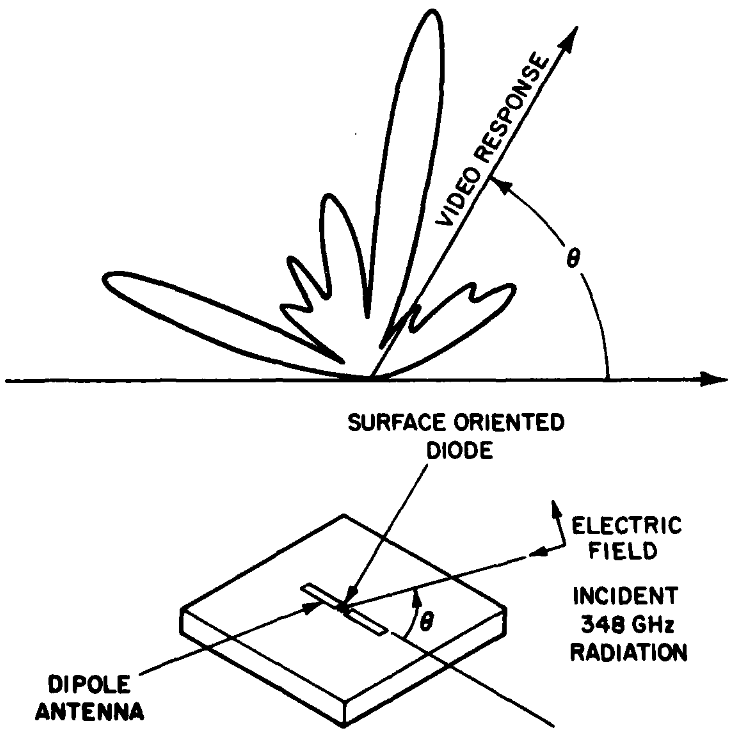

Fig. 6 Antenna pattern of planar diode with attached half-wavelength dipole.

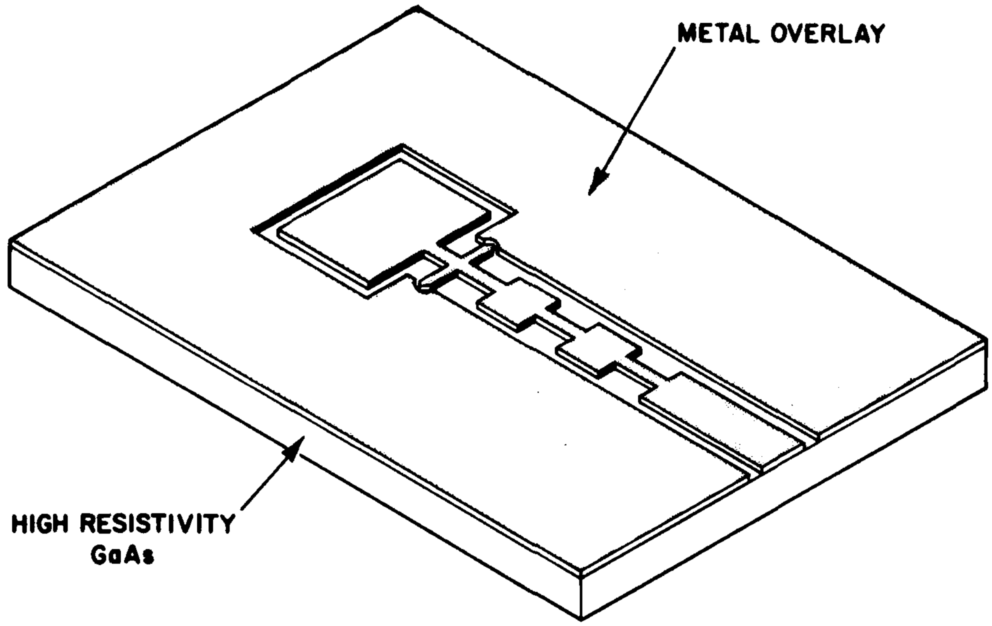

Fig. 7 Model of monolithically integrated planar mixer diode and slot antenna.

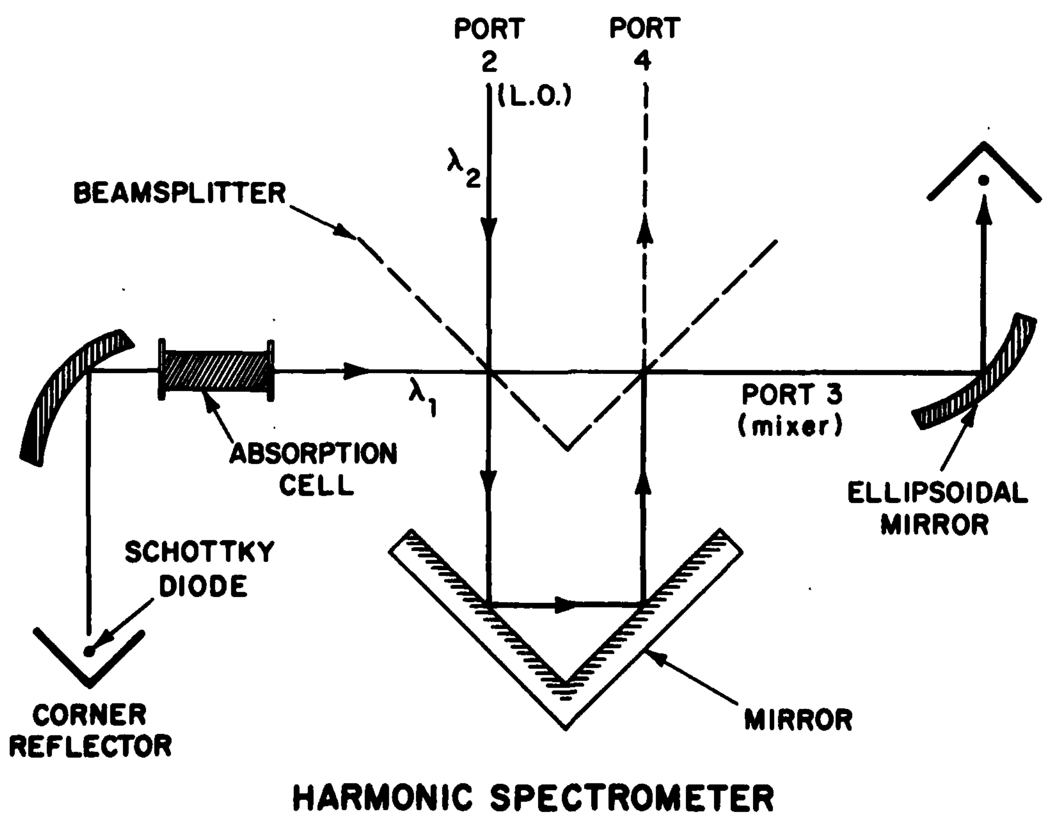

Fig. 8 Experimental high resolution spectrometer using quasi-optical diplexer and two Schottky diode corner reflectors.

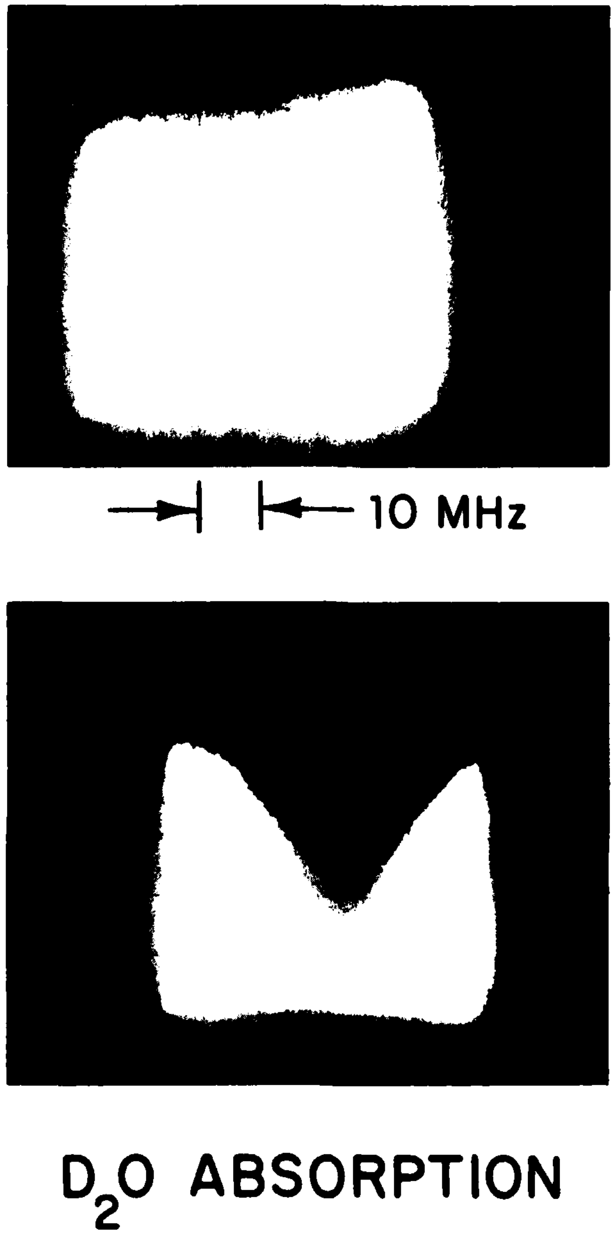

Fig. 9 Photographs of spectrum analyzer at 3 GHz showing the transmission of an absorption cell with (below) and without (above) D_2O gas.

Tunable submillimeter sources applied to the excited state rotational spectroscopy and kinetics of CH_3F [a)]

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Tunable submillimeter radiation, generated and detected using optically pumped lasers and Schottky diode mixers, has been used in an infrared-submillimeter double resonance investigation of CH_3F. This technique permits the direct observation of the molecular rotational spectra and kinetics of excited vibrational states and is particularly important for those molecules which are candidates for optically pumped submillimeter lasers.

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We report here the application of submillimeter lasers and Schottky diode mixers to an infrared-submillimeter double resonance investigation of the rotational spectra and kinetics of molecules in vibrationally excited states. There have been extensive infrared-microwave and infrared-infrared investigations of molecules.[1,2] Now for the first time, a broadly tunable *submillimeter* source has been used to probe directly excited state rotational levels which were heretofore inaccessible. The technique was applied to several sets of rotational energy levels in the ν_3 vibrationally excited state of CH_3F. In addition to demonstrating the utility of the method, improved values of particular molecular constants for the ν_3 state of CH_3F were obtained. We have also shown that the time dependence of population differences between pairs of rotational levels in the excited state could be observed with our tunable sources. This double resonance technique has many applications to excited state rotational spectroscopy and kinetics.

In these experiments the tunable submillimeter radiation was generated by nonlinear mixing in a corner reflector mounted Schottky barrier diode.[3,4] The output of an optically pumped submillimeter laser was mixed with a microwave signal to generate tunable sidebands. Conversion losses of the mixing process were sufficiently low that sidebands (with powers of up to 10^{-7} W) could be obtained using any of the large number of medium strength submillimeter laser lines. With a demonstrated sideband tuning range of ± 40 GHz, the density of usable laser lines is high enough to permit nearly complete coverage of the frequency region 300–900 GHz.

A tunable sideband spectrometer was constructed in which the tunable submillimeter radiation propagated through a sample cell and was coherently detected in a second Schottky diode mixer. The system is shown schematically in Fig. 1. To achieve high mixing efficiency, the output of the submillimeter laser was coupled into the diode by a diplexer[5] and an off-axis ellipsoidal mirror which focused the radiation into the main lobe of the antenna pattern of a long wire antenna mounted in a 90° corner reflector. The laser signal was mixed in the diode with a microwave signal introduced through the IF port of the diode, and the sidebands were reciprocally reradiated by the antenna structure. A diplexer separated the sideband and laser signals with low loss. The sideband beam entered and exited the 27-cm-long 2.2-cm-diam. sample cell through highly transparent natural quartz windows mounted at 45° to the axis of the cell. Coherent detection of the sidebands was effected in second corner

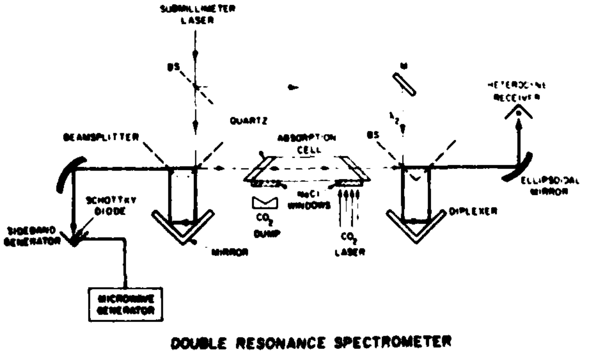

FIG. 1. Schematic diagram of the double resonance spectrometer. The intensity of the sideband beams transmitted through the sample cell was 40 dB above the receiver noise level.

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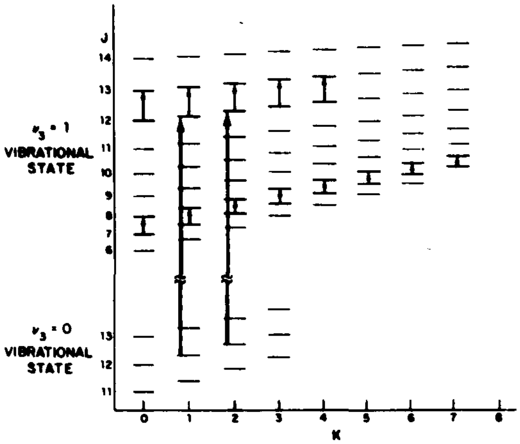

FIG. 2. Schematic energy level diagram of CH_3F showing the infrared vibrational transitions (dark arrows) pumped by the $9P(20)$ CO_2 laser line and the submillimeter rotational transitions (light arrows) observed in the cw double resonance experiment. J is the total rotational angular momentum and K is the component of J along the C-F-axis.

reflector mounted Schottky diode by mixing the sidebands with a portion of the submillimeter laser radiation to recover the microwave frequency signal. Then the microwave signal was amplified, filtered, and crystal detected. This heterodyne receiver was both very sensitive and fast.

To demonstrate the utility of the tunable sideband spectrometer, rotational transitions in vibrationally excited states were observed using an infrared-submillimeter double resonance technique. CH_3F was selected for investigation because of the importance of the CH_3F optically pumped submillimeter laser.[6,7] By driving ${}^qQ\,(12,K)$ rotation-vibrational transitions with the 9.55-μm $P(20)$ line of a CO_2 laser, certain $(J, K) = (12, K)$ rotational levels in the ν_3 vibrational state of CH_3F could be populated (where J is the total rotational angular momentum and K is the projection of J on the C-F axis). Radiative and collisional processes subsequently transferred population into the other low-lying (J, K) rotational levels of the ν_3 state. The $|\Delta J| = 1, \Delta K = 0$ rotational transitions, which are allowed by the electric dipole selection rule, could then be probed with the tunable sideband beam.

The CO_2 pump beam entered the sample cell through NaCl windows, reflected off the natural quartz windows, and propagated along the axis of the cell parallel or antiparallel to the submillimeter probe beam. The frequency of the CO_2 laser was tuned by optoacoustically monitoring the absorption of the pump beam with an electret microphone mounted in the cell. To observe excited state rotational spectra, the CH_3F was pumped wth a 15-W cw CO_2 laser. The pump beam was chopped at 500 Hz and the synchronous component of the transmitted intensity of the probe beam was detected. A pulsed CO_2 laser was substituted for the cw CO_2 laser to observe rotational kinetics. The CH_3F was pumped with 200-ns-wide 300-kw peak power pulses from a high repetition rate TEA laser which was grating tuned to maximum absorption by the CH_3F sample. The crystal-detected IF signal was sampled by a Biomation 8100-Nicolet Lab 80 data acquisition system. Electrical noise from the laser firing which was picked up by the Schottky diodes was greatly reduced by a background subtraction.

In the cw experiment, $(\nu_3, J,K) = (1,12,1)$ and $(1,12,2)$ levels were pumped and the line centers of the $J = 7\to 8$, $\Delta K = 0$, $K = 0,\ldots,7$, and the $J = 12\to 13$, $\Delta K = 0$, $K = 0,\ldots,4$ series of rotational transitions were measured, as shown in Fig. 2. To observe the $J = 7\to 8$ and the $J = 12\to 13$ series of transitions, tunable sidebands were generated by mixing the 742.6-μm (403721.6 MHz) and the 458.5-μm (653822.2 MHz)[8,9] laser lines of HCOOH with microwave signals from 500 to 1200 MHz.

At low pressures, the lineshape was highly asymmetrical only for the directly pumped $(J,K) = (12,2)\to(13,2)$

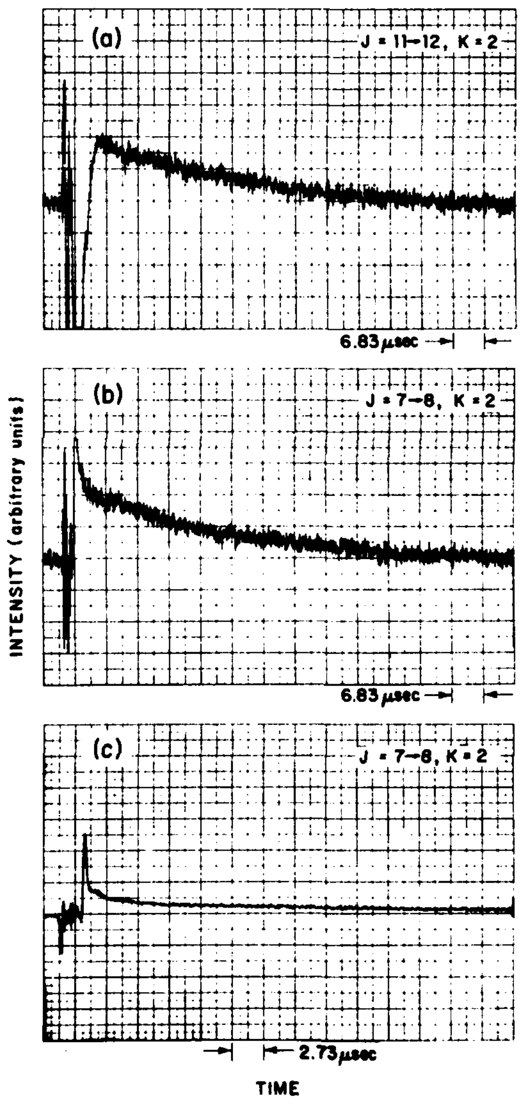

FIG. 3. (a) Stimulated emission pulse followed by absorption of the CH_3F submillimeter laser beam at a pressure of 25 mTorr. (b) Absorption of a sideband beam tuned to the $(\nu_3, J,K) = (1,7,2)\to(1,8,2)$ transition at a pressure of 1.19 Torr. (c) Superfluorescence pulse followed by absorption of a sideband beam tuned to the $(\nu_3, J,K) = (1,7,2)\to(1,8,2)$ transition at a pressure of 1.19 Torr.

transition. In this case, the CO_2 laser pumped a certain velocity group of molecules into the $(\nu_3, J, K) = (1,12,2)$ state, and velocity-dependent effects were seen. The collisions which populated the other rotational levels did not preserve the memory of which molecular velocity group was pumped. The tunable sideband spectrometer is well suited for detailed lineshape investigations of three and four level systems, and, in particular, the effect of the Raman-like processes in a three level system.[10-12]

The rotational energy of a symmetric top molecule such as CH_3 in the rotational state (J,K) and nondegenerate vibrational state $\nu_3 = v$ is given by

$$\begin{aligned}
&E_v(J,K)\\
&= B_v J(J+1) + (A_v - B_v)K^2 - D_{JKv} J(J+1)K^2\\
&\quad - D_{Jv} J^2(J+1)^2 - D_{Kv}K^4 + H_{Jv} J^3(J+1)^3\\
&\quad + H_{JKv} J^2(J+1)^2K^2 + H_{KJv} J(J+1)K^4 + H_{Kv}K^6,
\end{aligned}$$

which neglects, in particular, the perturbation of the ν_3 rotational levels by the ν_6 levels. The line centers of the $J = 7 \to 8$ and $J = 12 \to 13$ series of transitions were fitted to $E_v(J,K)$ for the rotational energies. The best fit to the line centers provided values of $D_{J1} = 0.0568 \pm 0.0005$ MHz and $D_{JK1} = 0.517 \pm 0.004$ MHz. Because of their relatively small magnitude, significant values of the H parameters were not obtained in this experiment. The measurements of D_{J1} and D_{JK1} lie within the error limits of the values obtained by Freund *et al.*[13] and the error limits of the present measurement overlap those from the fit by Arimondo *et al.*[14] of all data reported by 1978.

The principal source of error in the determination of the line centers was the reproducibility of the submillimeter laser frequency when the laser was returned to the peak of its gain profile. It was not possible to measure all the frequencies of the various $(J\text{-}1, K) \to (J,K)$ transitions for a given J before it was necessary to readjust the submillimeter laser. As a result, the standard deviation of the line center measurements varied from 0.2 to 0.8 MHz. No significant pressure shift was observed over the pressure range of 25–140 mTorr used in the line center measurements.

To demonstrate that the same mixing techniques could be used to investigate the kinetics and relaxation processes in the excited state, the CH_3F was pumped with the TEA laser. Three different types of mechanisms were observed: stimulated emission, collisional energy transfer, and superfluorescence. Stimulated emission was seen when the $(\nu_3, J, K) = (1,12,2) \to (1,11,2)$ transition was directly driven by the 496-μm CH_3F laser line. In this case, sidebands of the 496-μm laser line were used as the local oscillator to demodulate the transmission of the laser beam. A large stimulated emission pulse coincident with the pump pulse from the TEA laser was observed. A graph of the stimulated emission pulse followed by the exponential decay of the absorption of the submillimeter laser beam at a CH_3F pressure of 25 mTorr is presented in Fig. 3(a). Optical nutation[15] driven by the pump pulse was not seen since the Rabi flopping frequency was much higher than the 30-MHz bandwidth of the IF filters.

The exponential decay of the absorption of sideband beams was also observed. Figure 3(b) shows the decay of the absorption of a sideband tuned to the $(J,K) = (7,2) \to (8,2)$ transition at a pressure of 1.19 Torr. Decay rates of 30 to 52 ms^{-1} were observed for this transition over the pressure range 0.1–1.2 Torr. The largest contribution to the measured decay rates came from collisional redistribution of energy among internal degrees of freedom of the CH_3F. The contribution to the decay rates from both the known coupling of internal and translational degrees of freedom and diffusion of excited state molecules out of the probe beam was small.[16] The decay rates did not have a simple dependence on pressure and their interpretation was complicated by the fact that the TEA laser could pump several K levels of the ν_3, $J = 12$ rotational state. It is clear, however, that the use of low-pressure Q-switched pulsed lasers, which can populate a single K level, and Schottky diode reradiation techniques will permit the detailed analysis of the rotational kinetics of optically pumped molecules.

Superfluorescence directly from the $J = 12$ level was also observed. The intensity of the superfluorescence depended upon the CH_3F pressure and upon tuning the TEA laser to pump efficiently the CH_3F and did not depend upon the frequency of the probe beam. The effect of the superfluorescence pulse was to modulate the mixing efficiency of the diodes. Figure 3(c) shows a superfluorescence pulse followed by decay of the absorption of the sideband beam tuned to the $(J, K) = (7,2) \to (8,2)$ transition. The onset of the pulse was coincident with the pump pulse, and the width of the superfluorescence pulse was about 400 ns over the pressure range 0.1–0.6 Torr.

The reradiation of tunable submillimeter sidebands and their heterodyne detection, which has been demonstrated here, has many applications to molecular rotational spectroscopy and kinetics. The 300–900 GHz frequency region can be completely covered using known submillimeter laser lines. The reradiation technique is capable of very high precision since the Doppler limited linewidths of submillimeter frequency rotational transitions are a few megahertz and the source can be stabilized to within a few kilohertz. For molecules which can be optically pumped, the collision induced double resonance technique enables one to investigate both excited state rotational transitions which may be otherwise inaccesible and ground state rotational transitions with much improved sensitivity. Furthermore, the heterodyne receiver has been shown to be sufficiently fast to permit the observation of rapid time-dependent population transfer processes among rotational levels. The cross sections for collision induced rotational and rotation-vibrational transitions can be investigated for any molecule which can be optically pumped. In particular, this technique of double resonance using tunable submillimeter sources is especially appropriate for those molecules used in optically pumped submillimeter lasers.

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